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# SOVIET ATOMIC ENERGY

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# SOVIET ATOMIC ENERGY

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## ARTICLES

# RADIOPHOTOCHEMISTRY AS A POSSIBLE BASIS FOR THE EFFICIENT USE OF TWO-PURPOSE REACTORS

V. I. Gol'danskii

UDC 621.039.335

The recent extensive development of nuclear power has given rise to the extremely important problem of creating two-purpose reactors producing both electrical power and chemical products. Reactors of this kind may be considered with some justification as the basic installations of future large-scale radiochemical production [1-4]. A number of fundamental ways in which two-purpose reactors may be used for chemical production have been discussed in the literature, and these we shall now consider.

**Chemonuclear Synthesis.** This version is the most direct method of converting nuclear energy into the energy of endothermic chemical processes. The five stages involved in the trivial use of a nuclear reactor as an energy source for chemical production are replaced by a single stage (Fig. 1). The chief part of the fission energy — the kinetic energy of the fragments retarded in the reaction mixture (the proportion of fission energy carried away by the fragments  $f = 0.85$ ) — is in this case directly harnessed for chemical requirements. The mechanism underlying the conversion of the fission fragment energy into chemical energy in the gas phase is purely radiochemical; in the condensed phase this effect may be supplemented by the thermochemical action of the fragments [5, 6], associated with the formation of microscopic regions of brief local heating along the tracks of the fragments. The radiation yield of the transformations taking place under the influence of the fragments lies in the range  $G \sim 1-10$  for ordinary (not chain) reactions, where  $G$  is the radiation yield (the number of fully reacting molecules per 100 eV energy of the ionizing radiation), and differs little from that encountered in other forms of radiation [7].

The necessity of minimizing the primary radioactive contamination of the products of chemonuclear synthesis (even before subjecting these to special purification) compels us to use special highly dispersed chemonuclear elements or cells ("chels") as nuclear fuel; these have a certain optimum pore size, ensuring the liberation of a high proportion of the fission-fragment energy in the interior of the reagents, the fission fragments then emerging from the latter and being retained in the walls of the chemonuclear cell. This acts as a kind of self-filtration of the chemonuclear cell from radioactive contaminants [8-14]. The proportion of fission energy transferred to the reagents under optimum conditions is 0.3 [9].

Allowing for all these factors, the yield of chemonuclear-synthesis products for one act of fission of the nuclear fuel ( $E \sim 200$  MeV) is approximately  $2 \cdot 10^8 (G/100) f p \sim 5 \cdot 10^5 G$ . Here and subsequently we shall take the fixation of nitrogen ( $M = 28$ ) as a typical example; taking  $G = 1$ , we then obtain a yield of  $W_{\text{chn}} \sim 60$  kg/g U for the chemonuclear synthesis.

However, remembering that after the elapse of a time  $t \geq 10$  sec from the instant of uranium or plutonium fission the total activity of the fission fragments of 1 g of nuclear fuel equals  $3 \cdot 10^{11} t^{-1,2}$  Ci [15], it is easy to see that in the absence of further purification the activity of the chemonuclear synthesis products will be  $5 \cdot 10^6 t^{-1,2}$  Ci/g, i. e., in four or five days  $A_{\text{chn}} \sim 1$  Ci/g.

Requirements imposed upon the purification of chemonuclear-synthesis products are extremely strict (purifications of the order of  $10^9-10^{12}$  times are demanded); this makes the technology of chemonuclear processes more expensive and (even more important) makes them partially or even entirely incompatible with the elimination of pollution from the environment.

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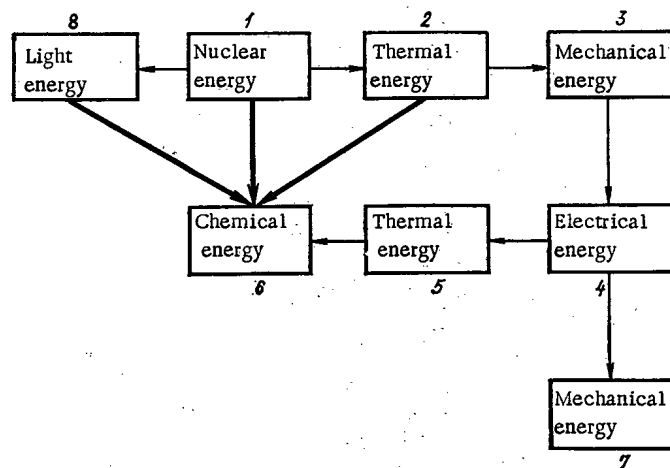


Fig. 1. Various ways of converting nuclear into chemical energy: 1-2-3-4-5-6) ordinary way; 1-6) chemonuclear synthesis; 1-2-6) high-temperature fuel element version; 1-8-6) radiophotochemical synthesis.

**Synthesis Using High-Temperature Reactors.** The two-stage conversion of nuclear energy into chemical energy via a thermal stage (Fig. 1) was proposed and examined in relation to the oxidation of nitrogen in [16]. Here it is essential to use high-temperature fuel elements; however, an extremely important advantage of this version is that there is no direct contact between the fission fragments and the reagents, so that the radiochemical purity of the products of the chemical processes is assured. The yield of combined nitrogen for fuel-element temperatures of 1900-2500°C is (according to the estimates of [16])  $W_{\text{temp}} \sim 200\text{--}360 \text{ kg/g U}$  ( $W_{\text{temp}}$  increases further in this case as the result of a chain reaction in the oxidation of nitrogen). A high yield of the desired products is achieved by the successful combination of high-temperature kinetics and the rapid cooling of the reaction products, i.e., the quenching of the favorable high-temperature equilibrium. Unfortunately at the present time there are no fuel elements capable of prolonged operation without failure at can temperatures of 2000°C or even lower temperatures. Chemical synthesis in high-temperature nuclear reactors thus remains for the time being a purely speculative version.

**Synthesis under the Action of Mixed  $\gamma$  Radiation from Nuclear Reactors.** Even in the case of special reactor constructions ensuring the maximum use of mixed  $\gamma$  radiation for chemical purposes, the power of these radiation sources is no greater than 1-6% of the thermal power of the reactor [1]. Hence the yield of the desired product is only  $W_{\gamma} \sim (2.5\text{--}15) \text{ kg/g U}$ , i.e., far lower than in chemonuclear synthesis.

The danger of radioactive contamination (activation) of the synthesized products of the radiochemical processes nevertheless remains (although to a less serious extent than in the case of chemonuclear synthesis); this contamination often includes radioactive isotopes of elements entering into the composition of the products which are incapable of being separated chemically.

Averaging the data presented in [1] we obtain the following relationship (typical for the case of mixed  $\gamma$  radiation), being the integrated dose of irradiation and the integrated thermal neutron flux:  $F/D \sim (2\text{--}3) \cdot 10^{14} \text{ neutrons/(cm}^2 \cdot \text{Mrad)}$ .

It is thus easy to see that the formation of every gram of the desired product in typical nitrogen- and hydrogen-containing systems in a field of mixed  $\gamma$  radiation with a radiation yield of  $G \sim 1$  will be accompanied by the accumulation of  $\sim 10^{-4} \text{ Ci}$  of  $^{14}\text{C}$  and  $\sim 2 \cdot 10^{-9} \text{ Ci}$  of tritium distributed between the original reagents and the radiochemical-synthesis products, in proportion to their contents of nitrogen and hydrogen molecules.

Although these activities are many orders of magnitude smaller than those encountered in chemonuclear synthesis, they are still quite unacceptable. It is clear that the large-scale manufacture of chemical products involves the creation of serious radioactive contaminants, the necessity of reliably separating these from the chemical products, and necessarily a high degree of purification of the original reagents from impurities liable to be severely activated by thermal neutrons (for example, halogens, sulfur, a large number of metals, and so on). All this may make the technology of chemical synthesis under the action of mixed  $\gamma$  radiation from nuclear reactors more expensive, so greatly limiting its practical value.

**Synthesis in Radiation Circuits.** According to the results presented in [4], the power drawn from reactors in the form of the activity of the radiation circuits may be estimated as  $\sim 1\%$  of the electrical power of the reactor, which amounts, for example, to  $\sim 10^8$  g-eq  $\cdot$  Ra for an electrical power of 100 MW or a thermal power of 100/K MW, where K is the thermoelectric energy-conversion efficiency. From this we may estimate the yield of end product as  $W_{\text{circ}} \sim GK \cdot 1.7$  kg/g U, which, although well below even the mixed  $\gamma$ -radiation output, nevertheless has a considerable qualitative advantage (the products contain no radioactive contaminants). Hence the use of radiation circuits is at the present time the most promising version of the radiochemical use of nuclear reactors [4].

**Radiophotochemical Synthesis.** One further possibility of two-stage transformation remains promising, that of nuclear  $\rightarrow$  light  $\rightarrow$  chemical energy (Fig. 1). In this version the fuel elements act as photochemical elements or cells ("fels"); the fission fragments are retarded in specially selected (solid or gaseous) scintillators, while the light so created is focused by a set of reflectors into the ends of glass-fiber or other light guides, which bring this light into the zone of photochemical transformation. The yield of the end product  $W_{\text{phch}} \sim 18,000 \Phi \alpha / E$  kg/g U, where  $\Phi$  is the fragment/light energy conversion coefficient (the so-called light yield), E is the energy of the light quanta in eV,  $\alpha$  is the quantum yield of the photochemical reaction, allowing for the incomplete focusing of the light into the photochemical zone (in the present case there is no need to concern oneself with stopping the fission fragments in the interior of the scintillator, so that the factor p encountered in  $W_{\text{chn}}$  is no longer required). There are certain scintillators for which  $\Phi$  reaches  $\sim 0.1$ , for example,  $\Phi \sim 0.08$  in NaI(Tl) and  $\Phi \sim 0.15$  in  $\text{CaI}_2(\text{Eu})$ , the value of  $\Phi$  not depending on the ionizing capacity of the particles but being the same for both  $\beta$  particles and fragments [17]. For  $E \sim 3$  eV and  $\alpha \sim 0.1$  we then have  $W_{\text{phch}} \sim 60$  kg/g U. Of particular interest for radiophotochemical synthesis are gaseous (rather than solid) scintillators of the xenon type, possibly with the addition of light converters for displacing the center of the scintillations from the ultraviolet to the visible region at pressures amounting to tens of atmospheres.

As in the case of synthesis with the aid of radiation circuits, the products of radio photochemical synthesis contain no radioactivity. The yield of these products may, in fact, be much greater than that of the circuit version, and (a factor of no mean importance) the perilous necessity of removing radioactive materials (activity carriers in the circuits) from the active zone of the reactor is avoided (this procedure is replaced by the collection and transmission of light).

Naturally it is desirable to ensure selectivity of the chemical transformation, and the preferential yield of a prespecified product. This may be done more easily in a photochemical transformation than in a radiochemical process, for example, by choosing the appropriate light converters so as to optimize the light of the radiophotochemical sources as regards intensity and spectral composition. We should take note of some important initial successes in the conversion of fission fragment energy into not simply monochromatic but also coherent light, involving the use of nuclear reactors to pump lasers [18, 19], the efficiency of the nuclear/laser energy conversion reaching several percent.\*

Of course there are a number of problems in which photochemical interaction cannot replace radiochemical processes (as in chemical transformations, or the modification of materials opaque to visible light or other radiations close to the optical region); however, there may also be processes in which radiophotochemical synthesis may prove to be the best method of using nuclear energy for chemical purposes and may provide a firm basis for the operation of the most economic two-purpose (power and chemical) nuclear reactors.

Investigations into the following problems of radiophotochemical synthesis are thus extremely vital:

1. The choice of systems ensuring the maximum conversion factor for the transformation of kinetic fission-fragment energy into light energy (it is desirable to obtain a monochromatic or bright-line light spectrum of photoradiation origin, the radiative pumping of lasers being of particular interest).

2. Provision for the most efficient method of collecting and transporting light from the active zone of the reactor (this requires, in particular, a high radiation resistance of the scintillator, which should clearly be of the gas type), and also the transportation of the whole system of light reflectors in the active zone and the initial section of the light guide from the active to the photochemical zone.

\*We are not here considering other aspects of interaction between laser and radiation-based processes, such as the possibility of increasing the radiation resistance of chemical compounds or improving the selectivity of radiolysis by using laser beams as a background to the irradiated substances [20, 21].

3. Choice of the best photochemical processes, with due allowance for the requirements of the chemical industry, the emission spectra of scintillators severely irradiated with fission fragments, and also the possibility of using various sensitizers and spectral converters.

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# EXPERIMENTAL DOUBLE-FUEL-ELEMENT CHANNEL FOR THE REACTOR OF THE WORLD'S FIRST NUCLEAR POWER STATION

M. G. Bul'kanov, V. A. Kurov,  
V. D. Lazarevskaya, V. G. Potolovskii,  
and V. S. Sever'yanov

UDC 621.039.5

The construction of new types of fuel elements requires that reactor tests be carried out on both the fuel components and on experimental half-scale fuel elements. The channel being described is intended for verifying, under the conditions of the reactor of the world's first nuclear power station, new technical solutions for the fuel elements of the Beloyarsk nuclear power station and the Bilibinsk nuclear heat and electric power plant. The positive experience accumulated on the basis of tests of unique experimental fuel elements has enabled an experimental channel to be constructed which, when loaded with fissile material, is identical with the regular-fuel-element channel and can be used in place of it.

The channel is designed for a regular cell of the reactor of the world's first nuclear power station and has the same subcoupling dimensions. In the construction of the channel, certain improvements are taken into account which were adopted in the channels of the Beloyarsk nuclear power station and the Bilibinsk nuclear heat and electric power station, and directed at increasing their efficiency.

The experimental channel (Fig. 1) consists of two fuel elements, descending and ascending tubes, installed in metal and graphite sleeves, which form a cylinder with diameter 64 mm for the graphite sleeve and 63 mm for the steel components. The length of the channel is 6.6 m.

The coolant from the conduit reaches the dispensing collector through a connecting tube and an inlet central connecting pipe, and enters the pressure chamber located in the lower part of the channel through two descending tubes with diameter  $12 \times 0.6$  mm. The coolant passes upwards, washing the inside surface of the fuel element tubes, from the chamber through spiral tubular compensators, which take up the temperature expansions between the descending and ascending tubes. Later, the coolant enters the collecting vessel and leaves the channel through a side connecting pipe. The dimensions of the fuel elements are:

TABLE 1. Results of Fuel Element Tests in the Experimental Channel

Cell	Cell designation	Date of insertion (1970)	Date of removal	Burnup, kg/t	Av. heat flow, $10^6$ kcal/m <sup>3</sup>	Temp. of fuel element cladding, °C
05-14	DTM-87	22.V	12.I.74	20,3	1,31	400
05-08	DTM-88	10.VI	12.I.74	18,5	1,13	380
05-18	DTM-90	6.IX	12.I.74	17	1,36	420
09-16	DTM-92	29.VI	12.I.74	17,1	1,56	430
12-19	DTM-93	10.VIII	12.I.74	16,4	1,16	390
08-15	DTM-95	10.VIII	12.I.74	18,1	1,7	440
11-12	DTM-96	3.X	12.I.74	17,2	1,3	400
09-18	DTM-86	21.V	15.III.72	6,5	1,5	430
04-11	DTM-85	21.V	7.X.73	17	1,23	390
01-12	DTM-84	21.V	7.IV.72	5,1	1,19	390
06-13	DTM-94	13.VII	6.IV.72	5,3	1,5	430
13-10	DTM-91	28.VI	7.X.73	13,1	1,1	380
08-19	DTM-89	18.VII	7.X.73	16,5	1,0	360
02-11	DTM-99	11.IX	Being tested	15	1,21	390
10-05	DTM-100	8.IX	Being tested	14,5	1,16	390
03-20	DTM-97	8.IX	6.IV.72	4,95	1,35	410
05-04	DTM-98	11.IX	Being tested	16,7	1,17	390

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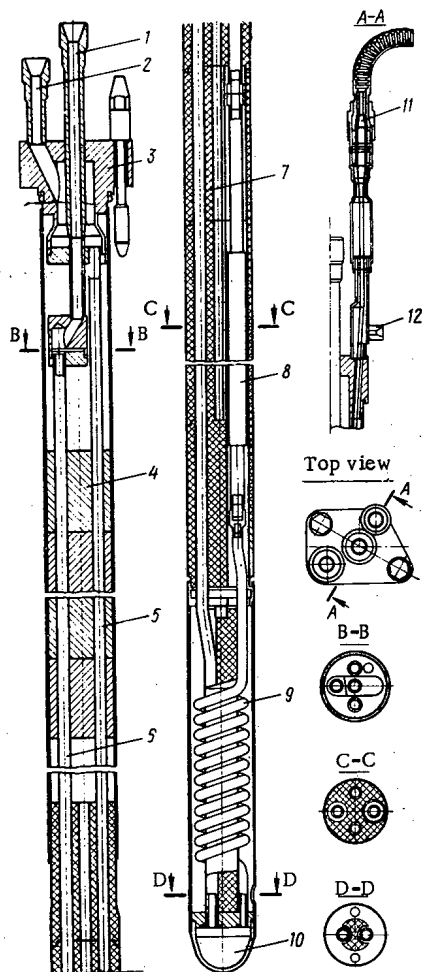


Fig. 1. Experimental double fuel element channel: 1, 2) inlet and outlet pipes for coolant of the first loop; 3) channel head; 4) steel sleeves; 5, 6) descending and ascending tubes; 7) graphite sleeves; 8) fuel element; 9) compensator; 10) rotary chamber; 11) thermocouple outlet assembly; 12) pressure bleeder pipe.

graphite sleeves are 0.5 to 0.7 mm (at the side). Heat from the graphite stack is removed largely through descending tubes, disposed in the graphite sleeves and with a minimum gap of about 0.15 mm. Part of the heat from the graphite is also transferred to the fuel elements.

In order to reduce the corrosive action of the reactor medium on the fuel-element cladding and tubes of the channel water circuit, provision is made for purging the channel with nitrogen. In order to ensure a reliable purging of the entire channel cavity, the compensation zone is enclosed on the outside with a tube of diameter  $63 \times 1.5$  mm, which has three openings 20 mm in diameter for outlet of the gas. Gas gaps are used also for monitoring the leak-tightness of the outer cladding of the fuel elements and the channel tubes, and this is effected by pumping out gas through the gaps and measuring its radioactivity. A connecting pipe, located in the head of the channel, is used for feeding in the gas. In channels with temperature measurement of the outer fuel-element cladding, this connecting pipe is located at the thermocouple outlet joint.

The principal technical data and operating conditions of the standard and experimental fuel channels are given below [1, 2]:

\* Channels of this design allow fuel elements with maximum external diameter of up to 25-26 mm to be tested in the regular reactor cells (diameter 65 mm).

diameter of the surrounding cladding  $20 \times 0.2$  mm and inside tube  $12 \times 6.6$  mm.\* The compensators are made of tubes with diameter  $9.4 \times 0.6$  mm and have a spiral pitch of 30 mm.

Below the distributing and collector vessels are assembled the metal plugs of the biological shield, with openings  $12 \times 0.6$  mm diameter below the tube. The lower plug is braced to the outside jacket, made from  $63 \times 1.5$  mm tubing. The jacket is connected with the channel head and is a load-carrying structure for the plugs of the biological shield. The lower part of the descending tubes and the fuel element are enclosed in graphite sleeves. The opening in the center of the sleeve, with diameter 10 mm, is designed for emergency withdrawal of the channel from the reactor cell. In the zone of the compensators, graphite sleeves are arranged for centering them.

In the experimental dual-fuel-element channel, with thermometry of the outside cladding of the fuel element, additional annular recesses are provided in the graphite sleeves for positioning the bracing units of the thermocouples and openings for their lead-out. A special packing gland is provided at the channel head for the thermocouple lead-out. There is also a fitting for the pressure bleeder.

Chromel - Alumel cable-thermocouples, type KTMS, were used, of size  $2 \times 0.06$  mm, and which had shown excellent efficiency. Measurement of the temperature of the thin-walled cladding of the fuel elements is one of the most complicated types of reactor thermometry. Most frequently, a bracing is used for the hot junctions of the thermocouples by means of clamping or bimetallic rings - this is a unique method of avoiding damage to the fuel-element cladding (Fig. 2). A considerable drawback of this method is the loss, in the course of time, of the clamping properties of the ring which leads to breakdown of the contact between the cladding and the thermocouple hot junction, and to the appearance of an error in the thermocouple readings.

The channel components are made from grade 12Kh18N10T stainless steel. For avoiding cracks, thin-walled tubes with diameter  $12 \times 0.6$  and  $9.4 \times 0.6$  mm were welded only with butt seams. In order to form the necessary space for the radial expansion of the fuel elements during swelling and for reliable monitoring of the leak-tightness of the fuel elements, the gaps between them and the

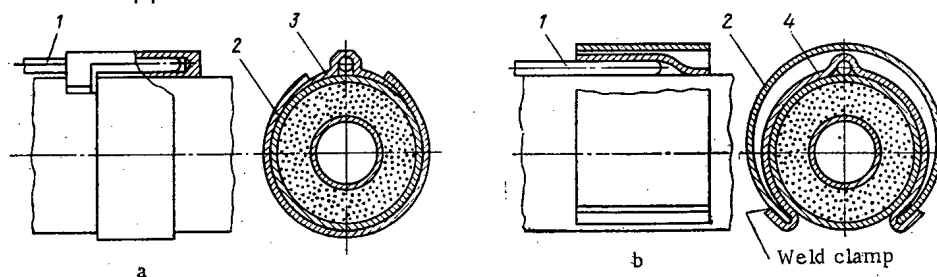


Fig. 2. Methods of securing the regulus of a thermocouple by means of a shoe (a) and a strip of foil (b): 1) thermocouple; 2) clamping ring; 3) shoe; 4) strip of foil.

Working pressure, kg/cm <sup>2</sup> .....	100, 100
Water temperature, °C .....	
at inlet.....	180-190, 180-190
at outlet .....	270-309, 270-309
Coolant flow rate, t/h.....	1.9-2.4; 1.2-1.5
Average thermal flow from fuel element to water, kcal/m <sup>2</sup> ·h.....	(0.8-1.8) · 10 <sup>6</sup> , (1-1.7) · 10 <sup>6</sup>
Maximum temperature of outer cladding of fuel element, °C .....	450, 450
Uranium charge in channel, kg.....	4, 4
Number of fuel elements, pieces.....	4, 2
Active length of fuel element, mm.....	1700; 1700
Outside diameter of fuel element, mm.....	14 × 0.2, 20 × 0.2
Diameter of central tube of fuel element, mm .....	9 × 0.4, 12 × 0.6
Quantity of steel per channel, cm <sup>3</sup> /cm.....	1.05, 1.2

The design of the channel permits the fuel elements to be tested under cavity boiling conditions, which is achieved when the channel is installed in the PV-2 experimental loop [2].

The operating parameters of the experimental channel under boiling conditions are shown below:

Pressure, kg/cm <sup>2</sup> .....	130
Water temperature at inlet, °C .....	250
Coolant flow rate, t/h .....	1.2
Channel power, kW.....	170
Steam content on exit from channel, % .....	7
Maximum temperature of outer fuel element cladding, °C.....	470

Tests of the channels under cavity boiling conditions were conducted with a steam content at the outlet of up to 7% by weight, which corresponded to the target of the experiment.

The main part of the experimental dual-fuel-element channels (see Table 1) was loaded into the reactor of the world's first nuclear power station in 1970. It can be seen from Table 1 that the fuel burnup in the fuel elements, the greater heat flows and the higher temperatures of the fuel element claddings confirm the operation of the experimental channels in more rigorous conditions by comparison with the standard fuel channels. The dual-fuel-element channels have operated in the reactor for about four years and are continuing to be used up to now. At present, fuel elements with heat exchange intensifiers are being tested in the experimental channels, with increased diameters and with a greater fuel charge.

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# STATISTICAL CHARACTERISTICS OF THE TEMPERATURE FLUCTUATIONS IN A DIRECT-FLOW SODIUM - WATER STEAM GENERATOR

B. V. Keadze, V. S. Sroelov,  
B. V. Kul'pin,\* and A. I. Gavrilin

UDC 621.039.534.63:621.039.534

The temperature fluctuations in heat-transfer systems produce thermal stresses and fatigue damage on prolonged operation. Considerable temperature fluctuations can arise in steam boilers at the start of the zone of deteriorating heat transfer on account of the alternating contact with liquid and steam [1]. The turbulent flow of the heat carrier in the presence of a considerable temperature gradient also produces temperature fluctuations which penetrate into the wall [2]. Finally, temperature fluctuations can also arise to some extent on account of the stochastic nature of the boiling, with random production and detachment of bubbles at the heated surface. Although there have been several studies of temperature fluctuations, it is not at present possible to estimate in advance the magnitude of the fluctuations for a particular apparatus, mainly on account of the difficulty of incorporating thermal and hydraulic characteristics of any particular design.

The basic purpose of statistical experiments is to determine the sources of the pulsations, the amplitude characteristics, and the characteristic times. The measurements were made with a model of a steam boiler using two standard modules made in Czechoslovakia. The body was  $159 \times 7$  m and length 6 m, which contained 19 tubes of diameter  $8 \times 3$ , which were fixed in double tubular assemblies. The thermal power of the testbed was 600 kW, but this did not allow us to produce normal mass flow speeds for the water in the 19-tube module. We therefore left only six working tubes, which lay at the edge of the tube bundle. The other tubes were blanked off and served for monitoring purposes. This enabled us to make measurements at flow rates in the range 400-850 kg/sec-m<sup>2</sup>, but the sodium speed in the generator was below the nominal value by about a factor of three. The tubes were made of NT8Kh6 steel. We mounted 50 microthermocouples in stainless-steel jackets of outside diameter 0.5 mm on one working tube at intervals of 190 mm in the

evaporator module near the outer surface of the body. The thermocouples were contained in longitudinal slots of depth 1 mm and introduced through the body. In the space between the tubes of the evaporator module, along an internal generator, there were 17 thermocouples in stainless-steel jackets of outside diameter 0.8 mm. Figure 1 shows the thermocouples used in most of the statistical experiments. The sodium flow rate was measured by an electromagnetic flowmeter calibrated to 2%. The water flow rate was determined from the pressure difference with a standard throttle using a differential manometer DM of class 1.5.

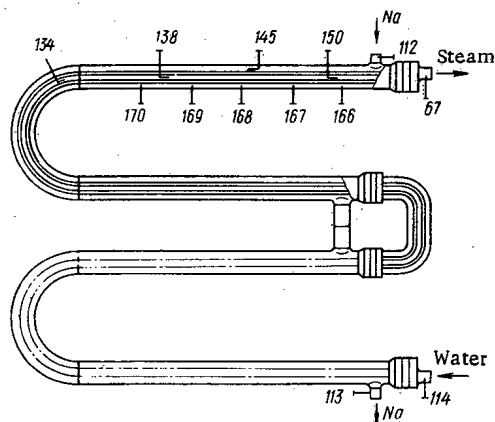


Fig. 1. The steam generator with transducers.

In each working state (Table 1) we measured the temperature distribution in the sodium and at the tube wall along the length of the boiler, which went with the input and output parameters of the sodium and water (steam) to provide a basis for calculating the heat fluxes and heat-transfer coefficients, for comparison with theoretical values.

\*Deceased.

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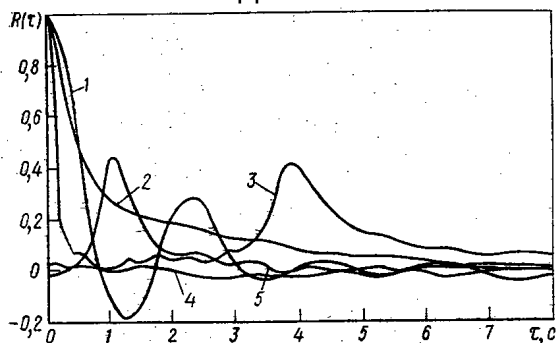


Fig. 2

Fig. 2. Autocorrelation functions for temperature fluctuations: 1)  $T_{138}$ ,  $\sigma = 4^\circ\text{C}$ ; 2)  $T_{134}$ ,  $\sigma = 1.4^\circ\text{C}$ ; 3)  $T_{134} - T_{145}$ ; 4)  $T_{167}$ ,  $\sigma = 2.4^\circ\text{C}$ ; 5)  $T_{166} - T_{167}$ .

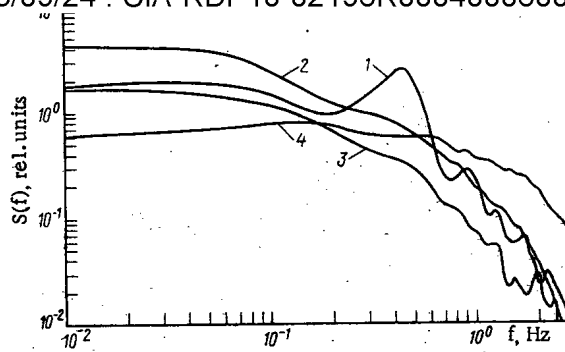


Fig. 3

Fig. 3. Spectral densities corresponding to the correlation functions of Fig. 2: 1, 2, and 4) as in Fig. 2; 3) modulus of the cross spectral density.

In the dynamic experiments, we measured primarily statistical characteristics of the thermocouple signals near the crisis region. We used electronic potentiometer recorders with 2-mV scales to record the temperature fluctuations.

The signals were amplified for statistical processing and converted to a pulse-frequency code for magnetic tape recording. This recording was performed simultaneously on 4 channels. Specialized computers (correlograph) calculated the mathematical expectation, dispersion, and autocorrelation functions, while a digital computer calculated the spectral densities. Pulsations of elevated amplitude occurred near the crisis region, which was first restricted to a length of 200 mm or so. The modes of operation were chosen so that the deviations of the thermocouple signal from the crisis zone to both sides of the mean were approximately equally probable. The pulsations remained stationary for 10-15 min. The precise mode of pulsation away from the crisis zone was less dependent on the working parameters.

In all we performed over 30 runs, in which we varied the flow rates, water temperature, pressure, and other parameters. We now consider some typical statistical characteristics. The normalized autocorrelation functions of Figs. 2 and 3 (except for curves 4 and 5) relate to the first mode (Table 1). The mode corresponds to a somewhat larger heat flux in the sections containing thermocouples 166 and 167. Thermocouples 134, 138, 145, and 150 were mounted in the tube, while 166-170 were in the sodium. The crisis zone lay near thermocouple 138. The fluctuations in the crisis zone had a standard deviation of about  $40^\circ\text{C}$  and contained considerable damp component with a period of about 2 sec, which was almost absent at adjacent thermocouples. The periodic form of pulsation was observed in many runs. Preliminary analysis showed that the reason may be some features of the water part of the system. For technical reasons, only part of the steam passed through a condenser and cooler, while the rest was mixed with the condensate directly ahead of the pump, which can lead to flow-rate fluctuations and temperature variations in the crisis zone. To test this, the fluctuations in water flow rate were written along with the other signals to the magnetic tape. We analyzed the normalized autocorrelation and cross-correlation functions for the flow rate and temperature in the crisis zone (Fig. 4, second mode, Table 1), and found a high (about 0.5) correlation coefficient, which was determined as

$$\kappa = \frac{|R_{xy}(r)_{\max}|}{\sqrt{R_{xx}(0)R_{yy}(0)}},$$

TABLE 1. Working Parameters of Steam Generators

$G_{\text{H}_2\text{O}}$ , tons/h	$W_0$ , kg/m <sup>2</sup> · sec	$t_{114}$ , $^\circ\text{C}$	$t_{113}$ , $^\circ\text{C}$	$G_{\text{Na}}$ , m <sup>3</sup> /h	$t_{112}$ , $^\circ\text{C}$	$t_{67}$ , $^\circ\text{C}$	$q_1^+$ , kcal/m · h	$q_1^-$ , kcal/m · h
1.08	445	252	341	12.2	462	304	41.7	147
1.25	512	287	333	9.4	508	306	51.4	208

Note. Steam pressure 100 atm;  $q_1^+$  and  $q_1^-$  linear heat fluxes before and after heat-transfer crisis zone.

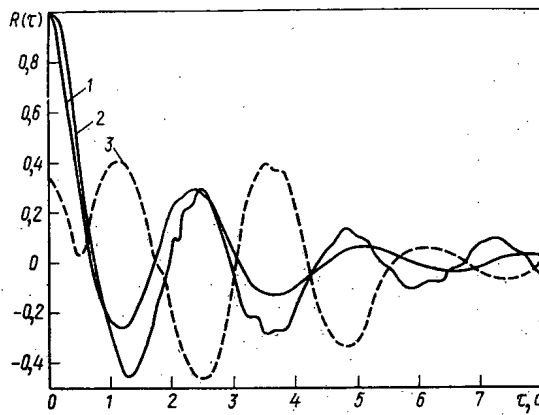


Fig. 4

Fig. 4. Autocorrelation functions for temperature fluctuations and water flow rate: 1)  $T_{138}$ ,  $\sigma = 6.3^\circ\text{C}$ ; 2)  $G_B$ ,  $\sigma = 0.6\%$ ; 3)  $T_{138} - G_B$ .

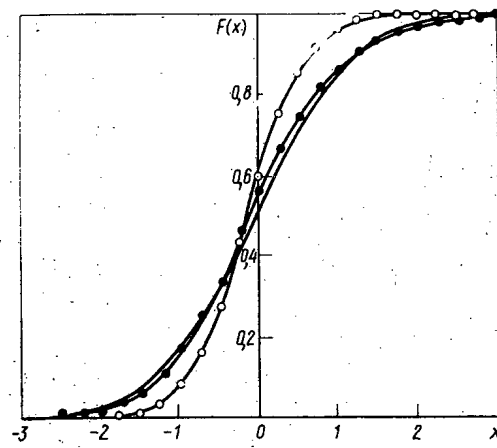


Fig. 5

Fig. 5. Distributions for the temperature fluctuations:  $\circ$   $T_{138}$ ;  $\bullet$   $T_{134}$ ; —) normal distribution.

as well as agreement between the periods and time shifts in the peak for the cross-correlation function, which indicates delay in the temperature with respect to the flow-rate variation. One can say that the latter variation is the external factor determining the character of the temperature pulsations. The measured relative standard deviations in the flow rate were small (maximal value about 1.2% in all runs), but the temperature fluctuations in the crisis zone became considerable ( $\sigma = 9^\circ\text{C}$ ), which corresponds to a doubled fluctuation amplitude of about  $25^\circ\text{C}$ . The amplitude of the temperature fluctuations increased as the frequency was reduced and as the amplitude of the flow-rate fluctuations increased. The temperature fluctuations in the crisis zone were anharmonic, as is clear from the content of higher harmonics in the spectral density (curve 1 of Fig. 3). The distributions (thermocouple 138, Fig. 5) differ substantially from normal, whereas the distribution for thermocouples outside the crisis zone was closely normal (thermocouple 134, Fig. 5).

The fluctuations in the thermocouple signals away from the crisis zone contained low-frequency and high-frequency components (curve 2 of Fig. 2). The periodic component was weak even when the water flow-rate fluctuations were present, for quite obvious reasons. The coefficient of heat transfer from the steam to the tube was low in the region of deteriorating heat transfer, and the thermocouple in the wall perceived in the main the fluctuations on the sodium side, whereas the thermocouple monitoring the steam at the output produced a periodic component correlated with the flow rate. In the section before the crisis region, the temperature in the tube was only slightly dependent on the flow-rate fluctuations.

We also used cross-correlation methods to detect the source of the temperature fluctuations in the tube away from the crisis zone. Figure 2 (curve 3) shows the cross-correlation function for thermocouples 134 and 145, which lie one on each side of the crisis zone; this contains almost solely a time-of-flight component, whose time shift agrees satisfactorily with the calculated transit time for sodium in that section. The same applies to the cross-correlation function for series of thermocouples in sodium (curve 5 of Fig. 2). Here the spectrum of the fluctuations was wider than that for the thermocouples in the tube (curve 4 of Fig. 2 and 3).

In measurements on the statistical characteristics of the sodium thermocouples it was found that the dispersion decreased away from the sodium inlet, and hence it was much lower in the lower part of the module. On the other hand, there was a marked rise in the dispersion on passing through the crisis region from the zone of deteriorating heat transfer to the region of dispersed annular flow, with the heat flux increasing by about a factor 4. The standard deviations for sodium thermocouples 166, 167, 169, and 170 were 3.3, 3.0, 1.9, and  $4.2^\circ\text{C}$ , respectively, when the crisis zone lay between thermocouples 169 and 170.

The relatively high-frequency fluctuations recorded by the thermocouples in the sodium and in the tube walls are due to motion of hydrodynamic nonuniformities of eddy type in the transverse temperature gradient. The strength of the eddies is particularly high where there is a destabilized flow near perturbation source (here the sodium inlet). The heat flux in the section between thermocouples 166 and 169 alters

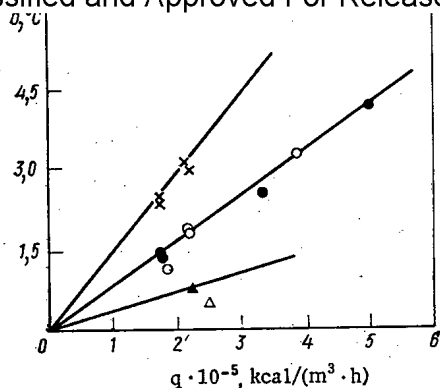


Fig. 6

Fig. 6. Standard deviation of temperature fluctuations in sodium as a function of heat flux for  $Re = \text{const} = 25,000$ :  $\times$ )  $T_{167}$ ;  $\circ$ )  $T_{169}$ ;  $\bullet$ )  $T_{170}$ ;  $\Delta$ )  $T_{181}$ ;  $\blacktriangle$ )  $T_{180}$ .

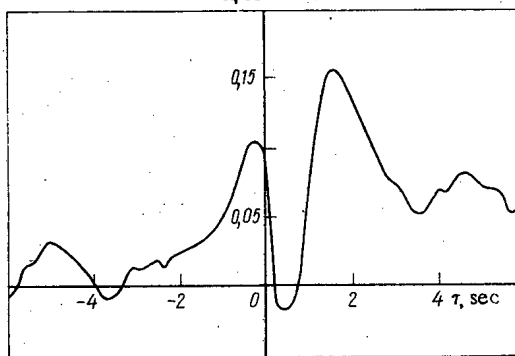


Fig. 7

Fig. 7. Correlation functions for the thermocouples in one cross section:  $T_{166}$  —  $T_{150}$ .

only slightly, and the decreased amplitude of the temperature fluctuations is due to damping of the hydrodynamic perturbation away from the sodium inlet. The increased pulsation near thermocouple 170 is due to the discontinuity in the heat flux. The sodium flow rate varied only slightly under the various conditions. The standard deviation of the temperature in the sodium as a function of heat flux is roughly of straight-line type (Fig. 6) for several thermocouples, and the fluctuations are larger for couples closer to the sodium inlet.

Figure 7 shows the cross-correlation function for thermocouples in the same cross-section at diametrically opposite edges of the body (in the sodium and tube). The sharp fall to negative values for small time shifts indicates that there are eddies comparable with the geometrical size of the steam-generator body ( $D_{\text{geom}}$ ). The large eddy size also explains the high correlation coefficient (0.4–0.5) for the thermocouples separated by a considerable distance along the axis ( $L/D_{\text{geom}} > 10$ ).

Then the temperature fluctuations in the wall in the crisis zone are largely determined by the flow-rate fluctuations; fluctuations in flow rate very minor from the operating viewpoint (about 1%) can result in temperature fluctuations of doubled amplitude up to 20–30°C. More detailed studies could be made of the relation between the flow-rate fluctuation and temperature fluctuation on a one-tube model.

Another appreciable source of temperature fluctuations lies in the large-scale eddies, which migrate in parts of destabilized flow due to the transverse temperature gradient. It is difficult to predict the exact extent of the fluctuations on a full-scale model on account of the differences in the temperature gradients and the hydrodynamic perturbations, but one should bear in mind that the level of the latter increases rapidly with the sodium flow rate. To reduce the importance of such fluctuations, one should avoid elevated temperature differences at parts where the flow is destabilized. In the present design, the zone of transition from deteriorating heat transfer to dispersed annular flow should be as far as possible from the sodium inlet.

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# ANALYSIS OF THE CONDITIONS REQUIRED FOR PRODUCING HIGHLY PURIFIED CARBIDE FUEL BY REDUCING OXIDES WITH CARBON, AND STUDY OF THE BEHAVIOR OF OXYGEN AND CARBON IMPURITIES IN THE PRESENCE OF FISSION PRODUCTS

R. B. Kotel'nikov, V. S. Belevantsev,  
S. N. Bashlykov, G. V. Titov,  
V. A. Zelyanin, and A. M. Anuchkin

UDC 621.039.542.394

The use of carbide fuel in fast reactors is one of several possible ways of achieving high economic efficiency [1, 2]. One of the most promising techniques for producing carbide fuel on an industrial scale is by reducing the oxides with carbon [3-5]. Certain questions associated with the production of carbide fuel by this method and with its subsequent use are still under discussion and require further investigation. This applies, in particular, to the problem of determining the optimum conditions for the production of carbide fuel with the minimum oxygen and carbon contamination and also the behavior of these impurities under conditions of irradiation.

A theoretical and experimental determination of the temperature corresponding to the onset of the reaction between uranium and plutonium oxides and carbon [6, 7] failed to produce the necessary agreement. We therefore calculated the temperature dependence of the oxygen potential in the U-C-O system by the Pourbaix method [8] using the latest thermodynamic data (Table 1) and plotted the corresponding diagram (Fig. 1). The free energy of formation of  $UC_{1-x}O_x$  was calculated from the equation

$$\Delta F = (1-x)\Delta F_{UC} + 4.576T(1-x)\lg(1-x) + x\Delta F_{UO} + 4.576Tx\lg x$$

on the assumption that ideal solid solutions of the UC-UO system were formed. The limiting solubility of oxygen in UC was taken as 17.5 at. % [12].

The minimum reaction temperatures obtained from the Pourbaix diagram for various pressures are given in Table 2, from which we see that the reaction underlying the production of uranium monocarbide by

TABLE 1. Thermodynamic Characteristics of Compounds Belonging to the U-C-O System

Compound	Equation for $\Delta F$ , cal/mole	Temp. range, °K	Ref.
CO(g)	$-26\,700 - 20.95\,T$	298-2500	[9]
UO <sub>2</sub> (s)	$-258\,000 + 40.00\,T$	298-1405	[9]
UO(g)	$-258\,300 + 40.50\,T$	1405-2000	[9]
UC(s)	$-121\,200 + 22.46\,T$	298-2500	[10]
UC <sub>2</sub> (s)	$-24\,260 - 1.12\,T$	298-1200	[11]
UC <sub>3</sub> (s)	$-25\,500$	1000-2200	[11]
UC <sub>0.65</sub> O <sub>0.35</sub> (s)	$-21\,300 - 3.00\,T$	298-2200	[11]
UC <sub>0.65</sub> O <sub>0.35</sub> (s)	$-22\,240 - 1.96\,T$	1100-1400	[11]
UC <sub>0.65</sub> O <sub>0.35</sub> (s)	$-24\,250 - 1.20\,T$	1400-2000	[11]
UC <sub>0.65</sub> O <sub>0.35</sub> (s)	$-58\,150 + 5.83\,T$	298-1200	[11]
UC <sub>0.65</sub> O <sub>0.35</sub> (s)	$-58\,950 + 6.56\,T$	1000-2200	[11]

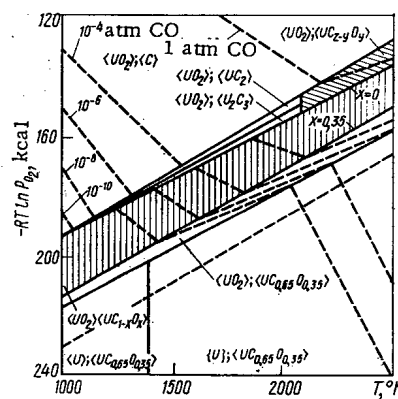


Fig. 1. Pourbaix diagrams for the U-O-C system.

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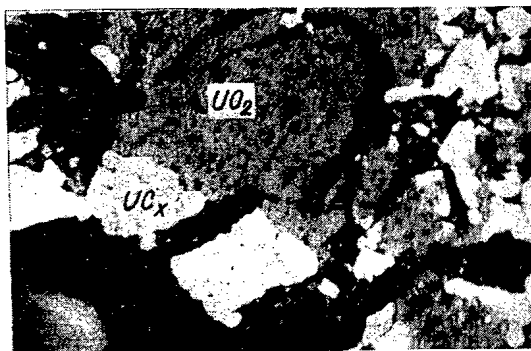


Fig. 2

Fig. 2. Microstructure of the reaction products after the interaction of uranium dioxide with carbon for 10 min at 1600°C.

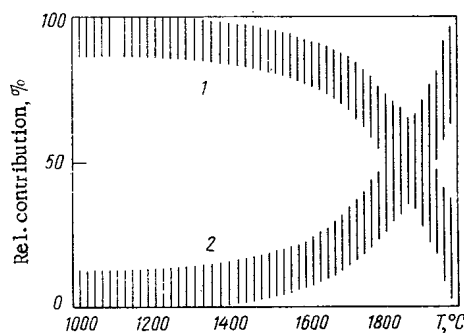


Fig. 3

Fig. 3. Relative contribution of various mass-transfer mechanisms in the carbiding of uranium dioxide: 1) diffusion; 2) evaporation — condensation.

the interaction of uranium dioxide with carbon may take place at comparatively low temperatures under the appropriate conditions.

The equilibrium quantity of oxygen dissolved in the uranium monocarbide depends on the temperature and partial CO pressure. According to Potter [13], the equilibrium quantity of oxygen in the product should be less than 0.05 wt. % for the conditions of producing uranium monocarbide by the carbothermal method usually employed in practice ( $T \approx 2000^\circ\text{K}$ , residual gas pressure  $10^{-3}$ – $10^{-4}$  mm Hg). The difficulties which arise in the practical synthesis of uranium monocarbide with such low oxygen contents are clearly associated with the mechanism of the reaction and with deviations of the composition and CO pressure from their average values within microscopic volumes of the charge.

In order to establish the mass-transfer mechanisms in the reaction of uranium dioxide with carbon, we studied the microstructure of the reaction products obtained under various conditions. As original materials we used uranium dioxide with a particle size of 2–10  $\mu$  and a specific surface of 3.4  $\text{m}^2/\text{g}$ , and various forms of carbon (soot, natural graphite, artificial graphite powder, etc.).

The powdered components were carefully agitated, converted into moldings, and heated in a vacuum of  $1 \cdot 10^{-4}$  mm Hg at 1600–2000°C for 10–20 min. The resultant products were studied metallographically; the analysis showed that the carbide phase was produced in the form of individual regions inside the uranium dioxide grains (Fig. 2); the shape and structure of the pores in the original uranium dioxide were preserved even after its transformation into the carbide; nowhere was any carbide phase found in the graphite particles, nor were there any continuous graphite-phase shells on the dioxide grains.

TABLE 2. Minimum Temperature Required for the Production of Uranium Carbides and Metallic Uranium as a Function of CO Pressure for the Interaction of Uranium Dioxide and Carbon

Partial pressure of CO		Max. reaction temp. ( $^\circ\text{C}$ ) with the formation of		
mm Hg	atm	UC <sub>2</sub>	UC	U
$7.6 \cdot 10^2$	1	1950	—	—
$7.6 \cdot 10^{-2}$	$10^{-4}$	1260	1480	—
$7.6 \cdot 10^{-4}$	$10^{-6}$	1140	1250	2270
$7.6 \cdot 10^{-6}$	$10^{-8}$	880	1050	1980
$7.6 \cdot 10^{-8}$	$10^{-10}$	770	930	1770

TABLE 3. Chemical Composition of Uranium Monocarbide Samples Containing Fission-Element Simulators

Element	Content, wt. %		Mean square error of the analysis, %
	in the charge	after anneal.	
U	88.90	88.0	1
C	5.3	5.3	2
Zr	0.75	0.70	1
Mo	0.92	0.90	0.5
Y	0.06	0.05	12
Ce	0.70	0.52	12
La	0.33	0.3	12
Nd	0.91	0.90	12
Pr	0.30	0.20	12
Sm	0.21	0.20	12
Ru	0.86	0.80	20
Pd	0.54	0.50	20
Sr	0.13	0.10	10
Ba	0.38	0.20	10
O	0.06	0.30	12
N	0.09	0.09	15
Fe	—	0.05	5



Fig. 4

Fig. 4. Microstructure of uranium monocarbide containing fission-element simulators ( $\times 500$ ): 1)  $\text{BaO} \cdot \text{SrO}$ ; 2)  $(\text{U}, \text{REE})_2\text{C}_3$ ; 3)  $(\text{REE}, \text{Y})_2\text{O}_3$ ; 4)  $\text{ZrC}$ ; 5)  $(\text{U}_{0.993}\text{Mo}_{0.007})\text{C}$ ; 6)  $\text{UMoC}_2$ ; 7)  $\text{U}(\text{Ru}, \text{Pd}, \text{Rh})_3$ .

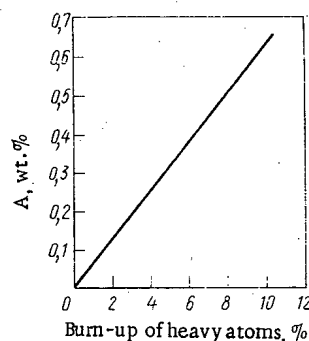


Fig. 5

Fig. 5. Amount of oxygen A combined by the fission-fragment elements (REE and Y + Ba + Sr) as a function of the depth of uranium monocarbide burn-up.

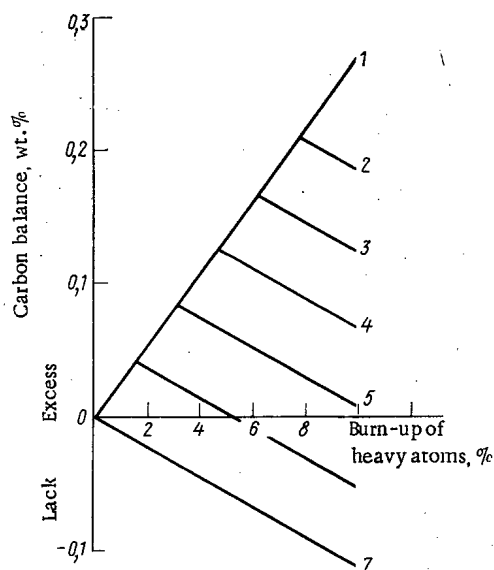


Fig. 6. Effect of the depth of burn-up on the carbon balance in uranium monocarbide for various initial oxygen contents: 1)  $\geq 0.64\%$  in original carbide 2)  $0.5\%$ ; 3)  $0.4\%$ ; 4)  $0.3\%$ ; 5)  $0.2\%$ ; 6)  $0.1\%$ ; 7)  $0\%$ .

These microstructural characteristics agree satisfactorily with the idea that the principal mass-transfer mechanism under the conditions in question was the solid-phase diffusion of carbon into the uranium dioxide. Signs of mass transfer by the evaporation of the dioxide, subsequent condensation, and interaction with carbon were only found at temperatures above  $1800^\circ\text{C}$  during the heating of pure uranium dioxide tablets in loose soot. Under these conditions a carbide layer was formed in the soot close to the upper end and lateral surface of the tablets (although with an appreciable gap relative to the dioxide tablet), while in the region of the lower end (at which the tablet was in close contact with the soot) the carbon clearly diffused into the uranium dioxide with the formation of a carbide layer. An approximate estimation of the contribution of the different mass-transfer mechanisms during the interaction of uranium dioxide with carbon in vacuo based on the results so obtained is presented in Fig. 3.

Under practical conditions of production the carbide fuel will of course contain a certain amount of oxygen and carbon, which (as indicated in a number of investigations) may have a marked influence on the chemical interaction with the fuel-element cans and the swelling of the fuel [10]. The behavior of oxygen and carbon impurities under conditions of intensive burn-up was studied by introducing fission-fragment simulators into the uranium monocarbide and then examining the composition of the phases. As original materials we used carbon thermally produced powders of uranium monocarbide alloyed with Zr, Mo, and Y, the hydrides of Ce, La, Nd, Pr, Sm, Ba, Sr, and metallic Ru and Pd. The amount of fission-product-simulating additives so incorporated approximately corresponded to a burn-up of 8-10% of the heavy atoms. Hot-pressing of the powder mixtures was carried out at temperatures up to  $1500^\circ\text{C}$ , after which the samples were annealed in hermetic molybdenum ampoules at  $1500^\circ\text{C}$  for 100 h.

All the operations of preparing the samples were carried out in an atmosphere of dry argon. We studied the chemical and phase compositions using the Cameca x-ray microanalyzer (Tables 3 and 4, Fig. 4).

TABLE 4. Results of a Study of the Phase Composition of UC Samples Containing Fission-Fragment Simulators (composition indicated in Table 3)

Name of phase	Microhardness, kg/mm <sup>2</sup>	X-ray microanalysis	
		composition of phase	conc. of ele- ments, wt. %
Uranium monocarbide with dissolved * molybdenum	1100±40	U Mo	90 Traces
Uranium sesquicarbide with dissolved REE and yttrium†	1550±30	U REE, Y	90 0,82
UMoC <sub>2</sub>	1400±30	U Mo	70 24
ZrC	2900±100	Zr	90
U (Ru, Pd) <sub>3</sub> (by analogy with the results of [14])	540±20	U Pd Ru	46 8 22
Solid solution of sesquioxides of REE and yttrium	—	REE, Y	80
Solid solution of barrium and strontium oxides	—	Ba Sr	Traces

\*Lattice constant 4.955 Å.

†8.093 ± 0.001 Å.

The results of our analysis of the phase composition agree closely with earlier published results [15, 16] and also with investigations into the composition of irradiated carbide fuel [14, 17]. It should further be noted that the rare-earth and alkaline-earth elements react with oxygen impurity, forming the corresponding oxides. The earlier observed fact that traces of the dicarbide originally present in uranium monocarbide vanish after irradiation [18, 19] is evidently due to the fact that the oxygen stabilizing the dicarbide has been combined with the fission elements.

The excess rare-earth elements (those remaining after interaction with oxygen) may then react with the excess carbon, forming sesquicarbides dissolving in U<sub>2</sub>C<sub>3</sub>. Ruthenium and palladium, and also, apparently, rhodium interact with the carbide, forming an intermetallic compound of the UMe<sub>3</sub> type, and increase the amount of active carbon by about 0.005% for each percent of uranium burn-up. Zirconium forms a carbide which under reactor conditions apparently dissolves in the carbide base. Most of the molybdenum takes part in the formation of a complex uranium molybdenum carbide.

On the basis of the foregoing scheme of chemical reactions between the fission products under consideration (Figs. 5 and 6) we estimated the changes taking place in the residual amounts of oxygen (dissolved in the monocarbide) and carbon (in the form of the dicarbide) during irradiation [20].

After determining the phase composition of the material simulating the UC-base fuel following the burn-up of 10% of the heavy atoms we calculated the theoretical density, which amounted to 13.4 g/cm<sup>3</sup>. This enabled us to obtain an approximate value for the solid swelling, which amounted to 0.4% for 1% burn-up.

### CONCLUSIONS

1. We have made a thermodynamic analysis of the U—C—O system by the Pourbaix method, and on the basis of the resultant diagram have determined the minimum temperatures required to produce uranium carbides and metallic uranium from uranium dioxide.

2. We have analyzed the various possible mechanisms for the mass transfer of the reagents in the production of carbides from oxides. The main contribution to mass transfer in the UO<sub>2</sub>—C system at temperatures up to 1800°C arises from the diffusion of the components through the layer of reaction products. The contribution of uranium dioxide evaporation to the mass-transfer mechanism becomes appreciable at temperatures above 2000°C.

3. We have studied the phase composition of hyperstoichiometrical uranium carbide samples containing fission-element simulators (Zr, Mo, Y, Pd, Ce, La, Nd, Pr, Sm, Ba, Sr, Ru) in quantities corresponding to 10% burn-up.

4. On the basis of the chemical reactions between the fragments under consideration we have estimated the changes taking place in the amounts of oxygen (dissolved in the monocarbide) and carbon (in the form of the dicarbide) during irradiation.

5. The calculated solid swelling of the carbide fuel was 0.4% swelling for 1% burn-up.

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# SIMULATION OF THE FORMATION AND ANNEALING OF DEFECTS IN REGIONS OF DAMAGE CREATED BY COLLISION CASCADES IN ALPHA IRON

P. A. Platonov, V. F. Krasnoshtanov,  
and Yu. R. Kevorkyan

UDC 620.19:539.2.001.57

Two computer programs have been created for simulating cascades of atomic collisions in cubic crystals and describing the isothermal annealing of point defects in regions of cascade-induced damage in  $\alpha$ -iron: these are known as SPIKE and HITANN. The SPIKE program realizes the cascade model on the approximation of paired elastic collisions. The thermal vibrations of the lattice atoms as well as the energy losses of the displaced atoms arising from inelastic interactions with electrons are not taken into account. The elementary act of interaction between the atoms is described by the equations of classical mechanics [1]. Calculation of the cascade is conducted by generations, the atoms in each generation being ordered with respect to velocity. This procedure enables us to allow for the overlapping of the various branches of the cascade, as a result of which a displaced atom is able to move into the regions damaged by atoms of earlier generations. A crystal atom is displaced from its lattice point if the energy imparted to it exceeds a certain threshold value. If the energy of the cascade atom falls below the threshold energy of the displacement, consideration of its history terminates, and it is thereafter interpreted as an interstitial atom (with the exception of substitutional collisions). The procedure for the localization of the interstitial atom employed in this program enables us to consider this atom as a possible target for an atom of a later generation passing through the same region. The cascade process ends when the energy of all the displaced atoms falls below the threshold. Then the damaged state is analyzed in order to estimate the recombination of the unstable Frenkel' pairs and the formation of complexes of defects.

Using the SPIKE program we studied the characteristics of the damaged regions created by cascades of collisions in  $\alpha$ -iron at 0°K. As interatomic interaction potential we used the Erginsoy — Vineyard version [2]:

$$U(r) = \begin{cases} \frac{0.7}{r} 8573 \exp(-6.547r), & r \leq 0.7; \\ 8573 \exp(-6.547r), & 0.7 < r < 1.35 \end{cases}$$

$r$  is expressed in units of half the lattice constant,  $U$  in electron volts). The displacement threshold equaled twice the energy of sublimation (8 eV). The recombination criterion was the stability of the Frenkel' pairs for  $\alpha$ -iron obtained by Vineyard's dynamic simulation technique [2]. According to these calculations an interstitial atom and a vacancy in  $\alpha$ -iron lying at a distance equal to the radius of the first, second, fifth, or tenth coordination spheres from one another recombine at 0°K, i.e., without thermal activation. In considering the formation of complexes, the assignment of a defect to a particular complex was estimated on the basis of the following criterion [3]: The distance between the defect and at least one vacancy of the complex should be no greater than the lattice constant (for a vacancy) or the distance to the nearest neighboring atom (for an interstitial atom).

We studied cascades initiated by first-ejected atoms in the energy range 0–25 keV. The average characteristics of the cascade regions of damage were found by analyzing a statistical set of 48 cascades for each energy of the first displaced atom. The random parameter in the statistical set of cascades was the direction of the velocity vector of the first-ejected atom, with a probability density distributed uniformly in space.

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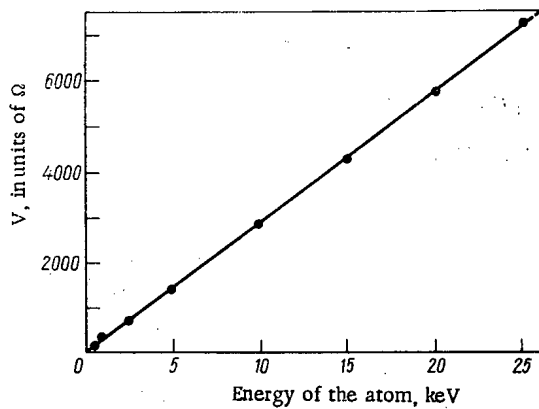


Fig. 1

Fig. 1. Volume of the cascade region of damage as a function of the energy of the first-ejected atom ( $\Omega$  is the atomic volume).

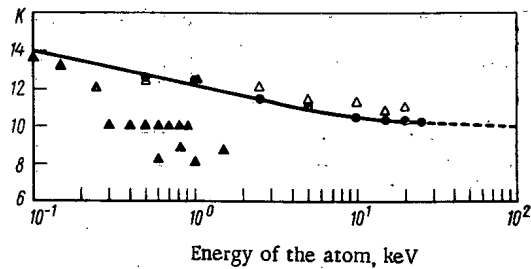


Fig. 2

Fig. 2. Efficiency of the displacement  $K$  as a function of the energy of the first-ejected atom:  $\bullet$  SPIKE;  $\Delta$  [3];  $\blacktriangle$  [5].

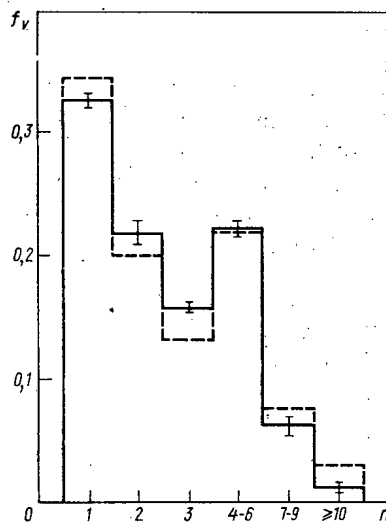


Fig. 3. Relative distribution of the total number of vacancies with respect to complexes of dimensions  $n$ :  $\longrightarrow$  SPIKE;  $\dashrightarrow$  results of [3].

The main characteristics of the cascade region of damage were the volume of the region, the displacement efficiency, the spatial distribution of the defects, and the size distribution of the defect complexes.

The volume of the region was estimated from the number of collisions in the cascade. It was assumed that the collision excited the volume of one unit cell equal to  $2\Omega$  in the bcc lattice ( $\Omega$  is the atomic volume). Thus the volume of the cascade region of damage equals  $2N_c\Omega$  ( $N_c$  is the number of collisions in a cascade). We see from Fig. 1 that this increases linearly with the energy of the first-ejected atom. We found that the mean density of stable displacements in the damaged region for this estimate of the volume was almost independent of the energy (within the region under consideration), amounting to  $\sim 3.6\%$ . This practically corresponds to the saturation density, which may be estimated as  $1/V_0$  ( $V_0$  is the recombination volume equal to  $30\Omega$  in the present model).

Figure 2 shows the effective displacement  $K(E) = \nu(E)/E$  [ $\nu(E)$  is the cascade function] in relation to the energy of the first-ejected atom. The fall in  $K(E)$  with increasing energy of the first-ejected atom is due to the increase in the proportion of recombining pairs on account of the increase in the degree of overlapping of the cascade branches.

Channeling has no practical effect on the behavior of  $K(E)$ . We see from Fig. 2 that for energies of the first-ejected atom greater than  $\sim 10$  keV the value of  $K(E)$  is practically constant, which agrees with the calculations of Robinson and Torrens [4], in which  $K(E)$  was independent of the energy of the first-ejected atom over the whole range (0-100 keV). For comparison Fig. 2 also shows the results of Beeler [3] obtained by means of a model analogous to that described in the present investigation, and the results of the dynamic simulation method [5]. Beeler's data, obtained by averaging over 96 cascades for each energy of the first-ejected atom, agree closely with our present results. The use of the dynamic method in the range 0.1-1.5 keV gives a smaller number of stable defects than paired-collision models. It should nevertheless be noted that all the values of  $K(E)$  obtained by the dynamic method are the result of single calculations for first-ejected atoms moving in a direction of fairly low symmetry in the crystal, except for the value corresponding to  $E = 0.1$  keV (obtained by averaging over 49 directions).

In order to bring the data under consideration into agreement with the classical Kinchin — Pease formula [ $\nu(E) = (E/2) Ed$ ] we must replace the usual value of  $E_d = 25$  eV by  $E_{\text{eff}} = 45-50$  and  $50-55$  eV for the model of paired collisions and the dynamic method respectively. Thus the number of stable pairs of defects is approximately half that in the structureless models using the approximation of solid spheres.

Using the corresponding criteria for the assignment of defects to a single complex, we obtained the average size distribution of the complexes of vacancies and interstitial atoms (i. e., the distribution with respect to the number of defects in the complexes) in the cascade regions of damage created by first-ejected atoms with various energies. Analysis showed that the relative size distribution of the complexes was practically the same for all energies. This is apparently because the defect density in the cascade region of damage does not depend on the energy of the first-ejected atom. Figure 3 shows the distributions of vacancy complexes averaged over all the energies under consideration obtained in the present investigation as well as Beeler's data [3]. In the interstitial distribution, single interstitial atoms predominate (over 90%). The probability of the formation of a complex with more than three interstitial atoms is negligibly small.

The HITANN program is a modification and augmented version of the DAIQUIRI program developed by Besco [6] and intended for simulating the migration of interstitial atoms in the bcc lattice of iron. The main addition to the program is a section allowing for the migration of vacancies. Using the Monte-Carlo method the course of each point defect in the lattice is traced over a discrete sequence of time intervals. Simulation of isothermal annealing was carried out for temperatures of 300 and 800°K.

According to the calculations of Johnson [7], the migration energies of isolated vacancies and interstitial atoms in  $\alpha$ -iron are 0.68 and 0.33 eV at 300°K, respectively. The rate of migration of an interstitial atom is  $10^6$  times greater than that of a vacancy, so that it is reasonable to assume that in the initial stages annealing takes place exclusively by way of the migration of interstitial atoms and pairs of these (migration energy 0.18 eV). All the vacancies and vacancy complexes as well as complexes of interstitial atoms with more than two defects may be considered as stationary. The time interval  $t$  was chosen on the basis of the condition that the probability of an isolated interstitial atom executing a jump in time  $t$  should be equal to 0.1. The rate of migration of a pair of interstitial atoms at 300°K is 300 times greater than that of a single interstitial atom, so that in a time  $t$  it may execute 30 jumps.

At 800°K the rate of migration of an interstitial atom is  $10^2$  times greater than that of a vacancy and ten times smaller than that of a pair of interstitial atoms. In this case the program operates as follows: After the elapse of every period of  $100t$  (during which only the migration of interstitial atoms is considered) each vacancy is given an opportunity to execute one jump, with a specified probability of success.

The conditions of recombination and complex formation at 300°K were the same as in the cascade model already described; at 800°K the recombination volume increased to  $62 \Omega$  [8].

In the damaged lattice the motion of the defects was subject to the influence of the field of other defects and was thus mutually correlated. The effect of the surrounding defect field on the migration energy of vacancies and interstitial atoms was considered in [7, 8]. A miscellaneous set of defect configurations was examined, and on the basis of the resultant data the jump probabilities of mobile defects were calculated for various conditions of correlated motion [8]. These data were used in the present investigation.

As original data we used the distribution of point defects in the cascade regions of damage obtained by means of the SPIKE program. For an energy of the first-ejected atom equal to 5 keV we chose eight regions of damage, which were subjected to isothermal annealing at 300 and 800°K. At 300°K annealing was carried out for a period of  $1000t$  ( $t = 8.0 \cdot 10^{-11}$  sec). At 800°K both the interstitial atoms and the vacancies migrated for  $1000t$  ( $t = 2.9 \cdot 10^{-13}$  sec) (velocity ratio 100:1), then all the interstitial atoms were "frozen," and for a period of  $1000t_1$  ( $t_1 = 100t$ ) only vacancies were allowed to migrate. This procedure accelerated the analysis of the annealing process; it was justified because in a period of  $1000t$  practically all the interstitial atoms remaining mobile had passed to a considerable distance from the region of damage and took no part in the interaction processes. It is nevertheless important to note that as a result of the recombination of migrating vacancies and interstitial atoms from the stationary complexes the latter may pass into the group of mobile defects. We found that this occurred only rarely and had no major effect on the final result.

Simulation enabled us to obtain the average kinetic characteristics of the annealing process: the time dependence of the rates of recombination and complex formation and also the spatial distributions of the defects and the size distribution of the complexes after the set annealing time. Figure 4 shows the spatial distributions of defects in a typical 5-keV cascade region of damage, before and after annealing, in the form of projections on the  $Z = 0$  plane. As a result of annealing at 300 and 800°K about 50 and 75% of the defects were respectively recombined. Figure 5 shows the distributions of the complexes of interstitial atoms and vacancies with respect to size before and after annealing.

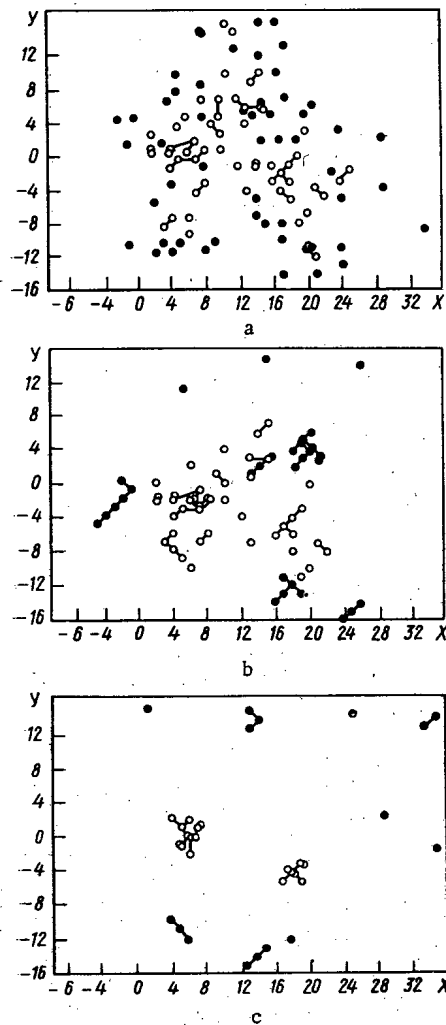


Fig. 4

Fig. 4. Cascade region of damage formed by a first-ejected atom with an energy of 5 keV (projection on the plane  $Z = 0$ ): a) before annealing; b and c) after annealing at 300 and 800°K, respectively; ○) vacancy; ●) interstitial atom; length dimensions  $1/2a$ , where  $a$  is the lattice constant (2.86 Å).

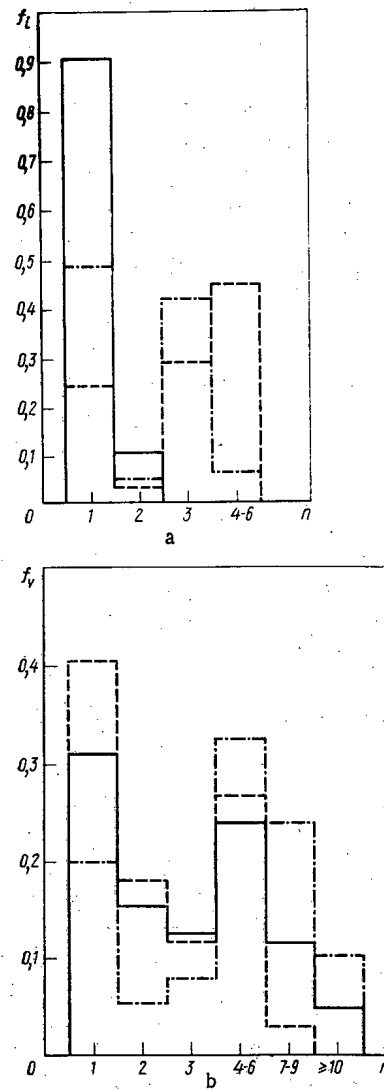


Fig. 5

Fig. 5. Relative distributions of the total number of interstitial atoms (a) and vacancies (b) with respect to complexes of size  $n$  (distributions averaged over eight 5-keV regions of damage): —) before annealing; ---, - · - · -) after annealing at 300 and 800°K, respectively.

The results of our present investigation are in excellent agreement with those of Doran [8], who used an analogous model to study the annealing of the cascade regions of damage obtained by Beeler [3].

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DETERMINING THE PROPORTION OF PLUTONIUM  
NUCLEI UNDERGOING FISSION DURING THE BURN-UP  
OF SLIGHTLY ENRICHED FUEL

K. I. Zykov and O. A. Miller

UDC 621.039.519:621.039.  
524.4:621.039.524.44

The extensive use of water-cooled, water-moderated reactors with slightly enriched fuel evokes interest in the nondestructive determination of  $^{239}\text{Pu}$  burn-up in irradiated fuel elements. Methods which may be used for this purpose include the  $\gamma$ -spectroscopy of the fission products. For the concentration of the fission product to depend on the  $^{239}\text{Pu}$  and  $^{235}\text{U}$  burn-up ratio, it is essential that the yields of this product arising from the fission of uranium and plutonium, respectively, should differ as much as possible. In order to determine the ratio of  $^{239}\text{Pu}$  and  $^{235}\text{U}$  nuclei undergoing fission, it was earlier proposed [1-5] that the concentration ratio of the  $^{106}\text{Ru}$  and  $^{137}\text{Cs}$  fragments should be used, since the  $^{106}\text{Ru}$  yield in the fission of  $^{239}\text{Pu}$  is an order of magnitude greater than in the fission of  $^{235}\text{U}$  (4.4 and 0.4%, respectively). For the ratio of the plutonium and uranium nuclei undergoing fission it is easy to derive the following relationship:

$$\frac{\Delta N_{239}}{\Delta N_{235}} = \frac{y_{106}^{235} - y_{137}^{235} \cdot \frac{(N_{106})_0}{(N_{137})_0}}{y_{137}^{239} \frac{(N_{106})_0}{(N_{137})_0} - y_{106}^{239}}, \quad (1)$$

where  $y_{106}^{235}$ ,  $y_{106}^{239}$ ,  $y_{137}^{235}$  and  $y_{137}^{239}$  are the  $^{106}\text{Ru}$  and  $^{137}\text{Cs}$  yields for the fission of  $^{235}\text{U}$  and  $^{239}\text{Pu}$ ;  $(N_{106})_0$  and  $(N_{137})_0$  are the quantities of the corresponding fission products arising from the fission of the nuclei.

In these measurements it was not the quantities  $(N_{106})_0$  and  $(N_{137})_0$  which were measured directly, but the number of  $^{106}\text{Ru}$  and  $^{137}\text{Cs}$  nuclei accumulating at the end of the irradiation period ( $N_{106}$  and  $N_{137}$ ), allowing for burn-up and decay. In order to obtain  $N_0$  from  $N$  it is essential to estimate the burn-up and decay of the fission products during the irradiation of the fuel. We may write

$$N_0 = NK, \quad (2)$$

where  $K$  is the corresponding correction for burn-up and decay. Since  $^{106}\text{Ru}$  and  $^{137}\text{Cs}$  are only formed in the direct fission of the fuel nuclei, the following expressions hold for the changes taking place in the concentration of each isotope:

$$\frac{dN(t)}{dt} = Q(t) - (\lambda + \sigma_a^N \Phi) N(t); \quad (3)$$

$$\frac{dN_0(t)}{dt} = Q(t); \quad (4)$$

$$Q(t) = [y^{235} N^{235}(t) \sigma_f^{235} + y^{239} N^{239}(t) \sigma_f^{239}] \Phi, \quad (5)$$

where  $\lambda$  is the isotope decay constant,  $\sigma_a^N$  is the cross section for the absorption of neutrons by the fission products,  $\sigma_f^{235}$  and  $\sigma_f^{239}$  are the neutron fission cross sections of  $^{235}\text{U}$  and  $^{239}\text{Pu}$  respectively,  $^{235}\text{N}$  and  $^{239}\text{N}$  are the concentrations of the isotopes undergoing fission,  $\Phi$  is the neutron flux density.

For  $^{106}\text{Ru}$  and  $^{137}\text{Cs}$  the neutron absorption cross sections are small (0.15 and 0.11b), so that the burn-up of these fission products may be neglected. Nevertheless, it is clear from the system of equations (2)-(5) that in order to calculate the correction factor we must know not only the irradiation time and the decay

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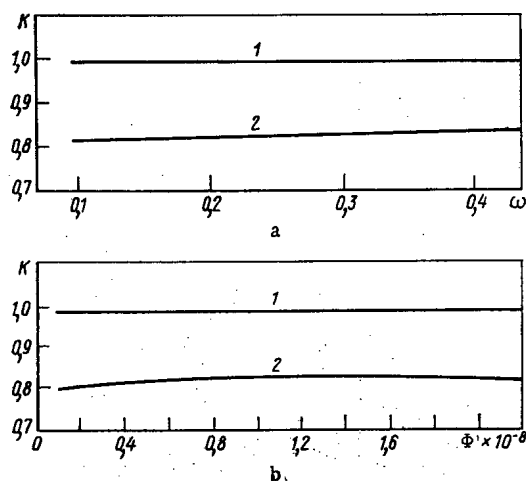


Fig. 1

Fig. 1. Dependence of the coefficient  $K$  of  $^{137}\text{Cs}$  (1) and  $^{106}\text{Ru}$  (2) on the parameter  $\omega$  and the quantity  $\Phi$ : a)  $\alpha = 0.226$ ,  $T_n = 550^\circ\text{C}$ ,  $t = 275$  days; b)  $\omega = 0.250$  and the same values of  $\alpha$ ,  $T_n$ , and  $t$ .

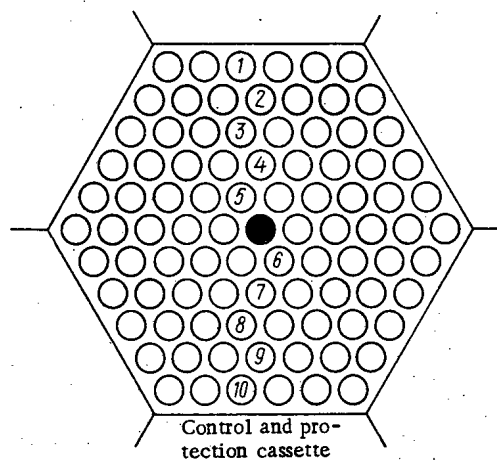


Fig. 2

Fig. 2. Arrangement of the fuel elements studied in the cassette: ●) central tube; 1-10) fuel elements studied.

constants but also the neutron flux and neutron cross sections of the isotopes undergoing fission. The neutron cross sections are determined by the neutron spectrum in the reactor. It was shown in [6] that any change in the neutron spectrum, even with respect to the height of a single cassette or from one fuel element to another, had a major influence on the accumulation of fission products for the same degree of fuel burn-up. Over the active zone as a whole this influence should be still greater, especially around the boundaries of the active zone, the absorbing elements of the control and protection system, and also in the case of cassettes differing substantially as regards uranium enrichment or burn-up.

Clearly for the same fission product  $N$  and  $N_0$  will depend in approximately the same way on the spectral parameters, and the correction factor  $K$  will be almost independent of them. Figure 1a shows the change taking place in  $K$  on increasing the resonance absorption coefficient in  $^{238}\text{U}(\omega)$  from 0.1 to 0.4 for  $^{106}\text{Ru}$  and  $^{137}\text{Cs}$ . The quantity  $\omega$  characterizes the number of captured neutrons (in the epithermal energy range of  $^{238}\text{U}$  in the volume of fuel under consideration) corresponding to the generation of one fast fission neutron in the same volume. Figure 1a shows that for  $^{106}\text{Ru}$   $K$  changes by no more than 2%, while for  $^{137}\text{Cs}$  it is practically independent of  $\omega$ . It follows from a detailed analysis of the computed data, including the influence of neutron temperature  $T_n$  and the ratio of the epithermal and thermal neutron fluxes  $\alpha$ , that for  $^{106}\text{Ru}$  the change in the correction factor  $K$  is no greater than 10% for any changes which may occur in the spectral parameters  $\omega$ ,  $\alpha$ , and  $T_n$  over irradiation times of between one and three years.

Figure 1b shows the dependence of  $K$  on  $\Phi = \sigma_a^{235} \varphi T$  ( $\sigma_a^{235}$  is the neutron absorption cross section of  $^{235}\text{U}$ ), from which it follows that in the present case  $K$  hardly depends at all on  $\Phi$  for  $^{106}\text{Ru}$  and  $^{137}\text{Cs}$ . Thus the correction factor for these fission products is mainly determined by the irradiation time and the half life of the product.

From Eqs. (2)-(5) we may derive the following equations if we neglect the burn-up of the fission product

$$K = g(t) \frac{\lambda t}{1 - e^{-\lambda t}}, \quad (6)$$

where

$$g(t) = \frac{e^{\lambda t} - 1}{\lambda t} \frac{\int_0^t Q(t) dt}{\int_0^t Q(t) e^{\lambda t} dt}. \quad (7)$$

A study of the expression for  $g(t)$  shows that this equals unity when  $Q(t) = \text{const}$ . This occurs if the isotope has an approximately equal yield for the fission of  $^{235}\text{U}$  and  $^{239}\text{Pu}$ , and if the reactor works at constant power. In any real cases the function  $Q(t)$  is not constant in time. For  $^{106}\text{Ru}$  the time dependence of  $Q(t)$  is quite strongly expressed in a reactor of the water-cooled, water-moderated type for a short time of irradiation. With increasing period of irradiation it becomes approximately constant.

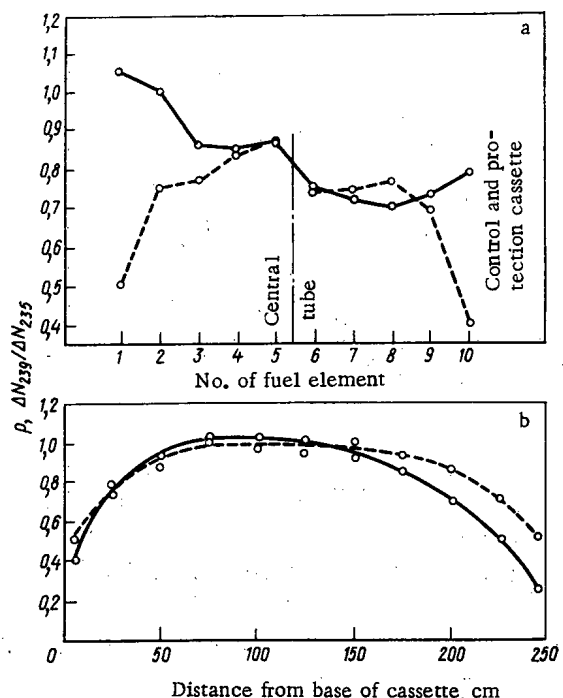


Fig. 3. Change in  $\Delta N_{239}/\Delta N_{235}$  (----) and fuel burn-up (—) over the diameter (a) and height (b) of the cassette p: a) the unit corresponds to a burn-up of 16.2 kg/ton and  $\Delta N_{239}/\Delta N_{235} = 0.517$ ; b) 18.2 kg/ton and 0.411, respectively.

calculated burn-up of 14.8 kg/ton, an initial uranium enrichment of 2%, and a holding period of 450 days. We measured 10 fuel elements arranged in the cassette as indicated in Fig. 2. A compensating cassette (control and protection system) was applied to one of the faces. All information regarding the characteristics of the fuel elements and cassettes was given in [7]. The concentration ratio of  $^{106}\text{Ru}$  and  $^{137}\text{Cs}$  was measured with a gamma spectrometer containing a Ge(Li) detector in the "hot" chamber of the Novovoronezh Nuclear Power Station. Each fuel element was measured at eleven points at different heights. The measuring method, procedure, and electronic apparatus were described in [4].

Using the measured concentration ratios of  $^{106}\text{Ru}$  and  $^{137}\text{Cs}$  and Eq. (1), we determined the ratio of the  $^{239}\text{Pu}$  and  $^{235}\text{U}$  nuclei undergoing fission. The correction factor was determined by means of the exact equations (2)–(5); the neutron flux and spectral parameters were calculated by the ROR program [7]. The error in determining  $f$  was  $\pm 2\%$ . Altogether the error in the proportion of  $^{239}\text{Pu}$  nuclei undergoing fission was  $\pm(12-13)\%$  and was mainly determined by errors in the nuclear data.

The mean value of  $\Delta N_{239}/\Delta N_{235}$  determined from the results of measurements on 110 points equaled  $0.396 \pm 0.051$ . The ratio of the  $^{239}\text{Pu}$  and  $^{235}\text{U}$  nuclei subject to fission depends both on the burn-up of the fuel (increasing together with the latter) and on the spectral parameters of the neutrons, increasing as the spectrum becomes harder (i.e., with increasing  $\alpha$ ,  $\omega$ , and  $T_n$ ). Figure 3a shows the change taking place in  $\Delta N_{239}/\Delta N_{235}$  and fuel burn-up over the diameter of the cassette. The burn-up distribution was determined from the change in the  $^{137}\text{Cs}$  concentration. The general downward slope in the distribution of burn-up on passing from fuel element 1 to 10 is due to the diminution in neutron flux close to the control and protection absorber, while the increase in burn-up in the extreme fuel elements is due to the rise in the thermal neutron flux  $\phi_T$  attributable to the effects of the water gaps between the cassettes.

It is an interesting fact that, despite the fall in burn-up from fuel element 1 to 5 and 10 to 8, the contribution of  $^{239}\text{Pu}$  increases in the corresponding directions. This is evidently due to the hardening of the neutron spectrum in the center of the cassette. For example, a change of this kind in the flux ratio of the epithermal and thermal neutrons  $\alpha$  over the cassette diameter resulting from the influence of water gaps and the surrounding cassettes was demonstrated in [5]. Since the number of fuel elements directly forming the faces of the cassettes amounts to one third of the total number of fuel elements, the influence of the

On the other hand, if the irradiation time is short compared with the half-life period, then  $\lambda t$  is a small quantity and  $g(t) \approx 1$ . In view of this,  $g(t)$  even for  $^{106}\text{Ru}$ , with its sharply expressed time dependence of  $Q(t)$  for short times  $t$ , differs little from unity for irradiation times of up to three years in neutron fluxes of up to  $10^{14} \text{ cm}^{-2} \cdot \text{sec}^{-1}$ . Thus for  $^{106}\text{Ru}$  and  $^{137}\text{Cs}$  we may use the simplified equation

$$K = \frac{\lambda t}{1 - e^{-\lambda t}}. \quad (8)$$

Detailed calculations of the quantity  $f = K_{106}/K_{137}$  based on the exact equations (2)–(5) and the approximate Eq. (8) show that the difference between these values is no greater than 15% for an irradiation time of between one and three years and any values of the burn-up and spectral parameters of the neutrons in reactors of the water-cooled, water-moderated type. For many practical cases an error of 15% is acceptable, so that there is no need to proceed to an exact calculation of  $f$ , for which it would be essential to know at least the approximate values of  $\omega$ ,  $\alpha$ ,  $T_n$ , and  $\Phi$  for every sample measured. The approximate formula enables us to determine  $f$  if we only know the fuel irradiation time.

The authors measured the proportion of  $^{239}\text{Pu}$  nuclei undergoing fission for a fuel cassette of the VVER-210 type in the first section of the Novovoronezh Nuclear Power Station, having a mean calculated burn-up of 14.8 kg/ton, an initial uranium enrichment of 2%, and a holding period of 450 days.

water gaps between the cassettes on the average value of  $\Delta N_{239}/\Delta N_{235}$  should be very considerable.

Figure 3b shows the change in  $\Delta N_{239}/\Delta N_{235}$  and fuel burn-up over the height of the cassette. We see that in the upper part of the cassette the proportion of  $^{239}\text{Pu}$  nuclei undergoing fission diminishes more slowly than the burn-up of the fuel. It is reasonable to assume that this is associated with the influence of the control and protection absorbers, which during the operation of the reactor are gradually drawn out of the active zone in conformity with the burn-up of the fuel and the accumulation of slags. The influence of the absorbers manifests itself, in particular, as a hardening of the spectrum; thus the neutron spectrum averaged over the irradiation period will be harder in the upper parts of the cassette, and this will lead to a certain increment in the proportion of  $^{239}\text{Pu}$  nuclei undergoing fission.

The experimental results confirm the computer data as to the influence of the neutron spectrum in the cassettes on the burn-up (and hence the accumulation) of  $^{239}\text{Pu}$ .

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# TECHNOLOGICAL SENSITIVITY FACTORS IN ATOMIC ELECTRIC POWER PLANT OPTIMIZATION

A. M. Kuz'min

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Technicoeconomic indices for atomic electric power plants are determined where there occurs some scattering of the technological parameters such as data on the properties of materials, cost characteristics, nuclear constants, and similar parameters. Errors in the values of technical parameters bring about uncertainty in such important characteristics as the cost of generated electric power or the amount of fuel consumption. For this reason, error estimates are of great significance in the basic indicators for atomic electric power plants (and above all in efficiency criteria), and the development of requirements for precision of knowledge about the technological parameters.

These problems have been treated with regard to physical nuclear constants in [1, 2], from which it follows explicitly that to evaluate the error in some value  $F_0$  one must know the sensitivity factors  $\delta F_0 / \delta \lambda_q$ :

$$\frac{\delta F_0}{\delta \lambda_q} = \lim_{\Delta \lambda_q \rightarrow 0} \frac{\Delta F_0}{\Delta \lambda_q},$$

where  $\Delta F_0$  is the change in  $F_0$  caused by the deviation of the  $q$ -th parameter of the value  $\Delta \lambda_q$  from the nominal value  $\lambda_q$ .

Let us examine a computation of the coefficients  $\delta F_0 / \delta \lambda_q$  for the case where the efficiency criterion of an atomic electric power plant  $F_0(v_1, \dots, v_n; \varphi; \lambda_q)$  is determined from the solution of the optimization problem:

$$\left. \begin{aligned} \min F_0(v_1, v_2, \dots, v_n; \varphi; \lambda_q); \\ F_l(v_1, v_2, \dots, v_n; \varphi, \lambda_q) \leq \\ \leq F_l^{\text{tol}}(\lambda_q), \quad l = 1, 2, \dots, m; \\ \alpha_j(\lambda_q) \leq v_j \leq \beta_j(\lambda_q), \quad j = 1, 2, \dots, n, \end{aligned} \right\} \quad (1)$$

where  $v_j$  represents the control parameters that are changed by making constraints in the additional functionals  $F_l(v_1, \dots, v_n, \varphi; \lambda_q)$  and minimizations in the criterion  $F_0$ ;  $\varphi$  is the state variables (for example, neutron fluxes within the reactor) that are subject to known equations;  $F_l^{\text{tol}}(\lambda_q)$ ,  $\alpha_j(\lambda_q)$ ,  $\beta_j(\lambda_q)$  are the tolerance values that depend on the magnitude of  $\lambda_q$  of the  $q$ -th technological parameter in the general case.

We will introduce the auxiliary variables  $w_l$ ,  $\omega_j$ , and  $\omega_{j+n}$  which can assume any value, and turn from the inequalities ( $F_l \leq F_l^{\text{tol}}$  or  $\alpha_j \leq v_j \leq \beta_j$ ) to the equivalent equalities (respectively  $F_l + w_l^2 = F_l^{\text{tol}}$  or  $v_j + \omega_j^2 = \beta_j$ ,  $v_j - \omega_{j+n}^2 = \alpha_j$ ). Designating  $u$  the dimensionality vector  $m_1 + 3n$  with the components  $\{v_j; w_l; \omega_j; \omega_{j+n}\}$  and implying by the variables  $A_j(\lambda_q)$  [ $i = 1, 2, \dots, m; m = m_1 + 2n$ ] the values  $F_l^{\text{tol}}(\lambda_q)$ ,  $\alpha_j(\lambda_q)$ , and  $\beta_j(\lambda_q)$ , we formulate problem (1) in the form:

$$\left. \begin{aligned} \min F_0(u, \varphi, \lambda_q); \\ F_i(u, \varphi, \lambda_q) = A_i(\lambda_q), \quad i = 1, 2, \dots, m, \end{aligned} \right\} \quad (2)$$

where the equalities contain constraints in the additional functionals and the controls of problem (1).

Problems (1) or (2) are problems in the calculus of variations for determining the conditional extreme. It can be solved for atomic electric power plants with fast reactors by means of the ROKBAR [3] program

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complex. Let a solution be obtained for problem (2) with the nominal value  $\lambda_q^0$  of the  $q$ -th parameter, i.e., a vector  $\mathbf{u}_0$  is found such that the necessary optimization conditions will be satisfied:

$$\mathcal{H}_0(\mathbf{u}_0, \lambda_q^0) + \sum_{i=1}^m \gamma_i^0 \mathcal{H}_i(\mathbf{u}_0, \lambda_q^0) = 0; \quad (3)$$

$$F_i(\mathbf{u}_0, \varphi_0, \lambda_q^0) = A_i(\lambda_q^0), \quad (4)$$

where  $\mathcal{H}_i(\mathbf{u}_0, \lambda_q^0)$  is the vector denoting the efficiencies of the variables  $u_j$  in relation to the functionals  $F_i$  ( $i = 0, 1, \dots, m$ ) obtained by using the equations in perturbation theory [4] at the point  $(\mathbf{u}_0, \lambda_q^0)$ ; and  $\gamma_i^0$  are the Lagrange factors. Since functionals  $F_i$  of problem (2) do not depend simultaneously on all auxiliary variables  $w_l$ ,  $\omega_j$ , and  $\omega_{j+n}$  (for example, criterion  $F_0$  depends only on the control parameters  $v_j$ ), several of the components of the vectors  $\mathcal{H}_i$  equal zero.

If there occurs a deviation in the  $q$ -th technological parameter for the value  $\delta\lambda_q = \lambda_q' - \lambda_q^0$ , we are confronted with a perturbed problem whose solution yields new values for the optimum vector  $\mathbf{u}' = \mathbf{u}_0 + \delta\mathbf{u}$ , state variables  $\varphi' = \varphi_0 + \delta\varphi$ , and efficiency criterion  $F_0(\mathbf{u}', \varphi', \lambda_q') = F_0(\mathbf{u}_0, \varphi_0, \lambda_q^0) + \delta F_0$  while keeping the constraints

$$F_i(\mathbf{u}', \varphi', \lambda_q') = A_i(\lambda_q'). \quad (5)$$

Considering the deviation  $\Delta\lambda_q$  rather small, we have

$$\Delta F_i = \frac{\partial F_i(\mathbf{u}_0, \varphi_0, \lambda_q^0)}{\partial \lambda_q} \cdot \Delta\lambda_q + \{\mathcal{H}_i(\mathbf{u}_0, \lambda_q^0) \Delta\mathbf{u}\}; \quad (6)$$

$$\Delta A_i = \frac{\partial A_i(\lambda_q^0)}{\partial \lambda_q} \Delta\lambda_q, \quad i = 0, 1, \dots, m,$$

where the expression  $\{\mathcal{H}_i \Delta\mathbf{u}\}$  designates the scalar product of vectors  $\mathcal{H}_i$  and  $\Delta\mathbf{u}$ . Using Eqs. (3)–(6) and assuming the existence of the continuous derivatives  $\partial\mathbf{u}/\partial\lambda_q$  and  $\partial\gamma_i/\partial\lambda_q$ , we obtain the equation

$$\frac{\delta F_0}{\delta \lambda_q} = \frac{\partial F_0(\mathbf{u}_0, \varphi_0, \lambda_q^0)}{\partial \lambda_q} + \sum_{i=1}^m \gamma_i^0 \left[ \frac{\partial F_i(\mathbf{u}_0, \varphi_0, \lambda_q^0)}{\partial \lambda_q} - \frac{\partial A_i(\lambda_q^0)}{\partial \lambda_q} \right], \quad (7)$$

in which the partial derivatives  $\partial F_i/\partial\lambda_q$  ( $i = 0, 1, \dots, m$ ) denote the efficiencies of parameter  $\lambda_q$  to the functionals  $F_i$ .

The factors  $\gamma_i^0$ , which are necessary for calculating the sensitivity factors  $\delta F_0/\delta\lambda_q$ , are determined either by solving from system (3)  $m_1 + 3n$  linear equations with  $m = m_1 + 2n$  unknowns, or by solving the duality problem in linear programming [5] for the variables  $y_i$  ( $i = 1, 2, \dots, m$ ). Actually, the best plan for the duality problem coincides with the Lagrange factors

$$y_i = \gamma_i^0 \quad (i = 1, 2, \dots, m),$$

if the correlations (3) are satisfied, while the basic problem of linear programming is formulated in the vicinity of the optimum control  $\{v_1^0, v_2^0, \dots, v_n^0\}$  for the variations  $\{\delta v_1, \delta v_2, \dots, \delta v_n\}$  (the same as in [6]). In this connection  $\gamma_i^0 = 0$  for  $i \in I_0$ , where  $I_0$  is the set of those indices which coincide with the constraints in problem (1) that assume a kind of rigorous inequality at the optimum point. We will note that a second method for obtaining the factors  $\gamma_i^0$  is conveniently used in the ROKBAR program, where the linear programming is solved by the method of successive reduction of discrepancies [5], which makes it possible to determine the optimum plans for the basic and duality problems.

It is important to stress that in the scheme proposed above, the sensitivity factors  $\delta F_0/\delta\lambda_q$  are determined for the optimum conditions of criterion  $F_0$  and there is no ambiguity in the calculation of  $\delta F_0/\delta\lambda_q$  which is associated with the ambiguity of the method used to regenerate possible violations of the constraints  $F_i = A_i$  (when the parameter  $\lambda_q$  is perturbed). It can easily be seen that one would choose in Eq. (7) among all possible methods (determined by the choice of equations  $v_j$ ) of recovering from the perturbed conditions the one method which produces the greatest lowering of the criterion  $F_0$ .

We will illustrate the practical use of Eq. (7) with the example of a calculation of the sensitivity factors of the doubling time  $T_2$  for fast-breeder reactors that use oxide fuel ( $\text{UO}_2 + \text{PuO}_2$ ) with respect to the limiting tolerances of the functionals  $F_i$ . Let the reactor have a heat power output of  $W_T = 1000$  MW and consist of a single core surrounded by screens containing reproductive material in the form of  $\text{UO}_2$ . We will consider the fuel to be contained in cylindrical fuel elements with stainless steel casing 0.4 mm in thickness. Liquid sodium at an average temperature of 530°C at the reactor outlet is taken as the heat

TABLE 1. Effectiveness of Functional Controls

Controls	Optimum values $v_0$	Efficiencies $\mathcal{H}_i^0$ of controls to functionals					
		$K_{\text{eff}}$	$t_{\text{ce}}, ^\circ\text{C}$	$t_{\text{ca}}, ^\circ\text{C}$	$\xi_{\text{ca}}$	$\sigma_w, \text{kg/mm}^2$	$T_2, \text{yr}$
Hg.p. cm	35,2	0	0	0	-0,03285	+0,0670	0
$\epsilon_{\text{h.w.}}$	0,114	-0,4177	+2355	+220,8	+0,01327	-315,6	-12,40
$R, \text{cm}$	88,1	+0,02194	-39,51	-3,704	-0,03218	0	+0,2048
$d_T, \text{cm}$	0,5	+0,1983	+5339	+57,43	+0,02788	-30,65	+6,315
$\Pi$	0,145	-0,7439	-3179	-21,95	+0,1273	0	-39,67
$h$	1,15	-0,6774	+3148	-633,1	-0,04512	-75,71	-24,07
$x$	0,163	+2,830	-20,20	-1,894	-0,03133	0	+164,7
$H, \text{cm}$	97,0	+0,02161	-17,50	-0,501	+0,04561	+0,0670	+0,1196
$V_{\text{h.e.}}, \text{m/sec}$	9,57	0	-0,1178	-16,03	-0,02102	+2,601	0
Factors $\gamma_i^0$		-58,28	+0,02151	+0,02810	+12,63	+0,0551	-

exchanger. We assume the chemical processing time for the fuel elements to be 0.5 year, and the thermo-physical and durability characteristics of the steel, fuel, and sodium to be the same as for the BN-350 reactor [7].

Let us examine the problem of the minimum doubling time  $T_2$  with the constraints

$$\left. \begin{aligned} K_{\text{eff}} = 1; t_{\text{ce}} \leq 2450^\circ\text{C}; t_{\text{ca}} \leq 710^\circ\text{C}; \\ \xi_{\text{ca}} \leq 0,002; \sigma_w \leq 18 \text{ kg/mm}^2 \end{aligned} \right\} \quad (8)$$

where  $K_{\text{eff}}$  is the effective neutron multiplication factor;  $t_{\text{ce}}$  and  $t_{\text{ca}}$  are the temperature maxima at the center of the fuel element and at its casing;  $\xi_{\text{ca}}$  is the upper limit of relative deformation of the casing; and  $\sigma_w$  is the maximum pressure at the fuel holder walls.

We will designate the following as the control parameters:  $v_1 = \text{Hg.p.}$  — the height of the zone for selecting fissionable gaseous products;  $v_2 = \epsilon_{\text{h.w.}}$  — the volumetric fraction of the holder walls;  $v_3 = R$  — the core radius;  $v_4 = d_T$  — brick fuel diameter;  $v_5 = \Pi$  — fuel porosity;  $v_6 = h$  — relative stage in the fuel element grating;  $v_7 = x$  — fuel enrichment;  $v_8 = H$  — height of core; and  $v_9 = V_{\text{h.e.}}$  — maximum rate of the heat exchanger. In this connection:

$$\left. \begin{aligned} 0 \leq \text{Hg.p.} < \infty; 5 \text{ mm} \leq d_T < \infty; 0 \leq x \leq 1; \\ 0 \leq \epsilon_{\text{h.w.}} \leq 1; 0,05 \leq \Pi \leq 0,174; 0 \leq H < \infty; \\ 0 \leq R < \infty; 1,15 \leq h < \infty; 0 \leq V_{\text{h.e.}} \leq 10 \text{ m/sec} \end{aligned} \right\} \quad (9)$$

The solution of this problem using the ROKBAR program with the basic assumptions about the computed reactor model that was described in [3] has shown that  $T_2^{\text{opt}} = 7.52 \text{ yr}$ ;  $t_{\text{ce}}$ ,  $t_{\text{ca}}$ ,  $\xi_{\text{ca}}$ , and  $\sigma_B$  attain the limiting tolerances, while the optimum controls are equal to the values given in Table 1. The table also presents the optimum control efficiencies  $\mathcal{H}_i^0$  and values of the factors  $\gamma_i^0$  obtained from the solution of the duality problem in linear programming.

Whereas problems of type (2) in the ROKBAR program are solved by approximation, substitution of the values found for  $\gamma_i^0$  under optimization conditions in Eq. (3) yields a difference from zero in the discrepancy  $\Delta_j = \mathcal{H}_j^0 + \sum_i \gamma_i^0 \mathcal{H}_i^1$ . It is easy to prove that the discrepancies  $\Delta_j$  are small also for each index  $j$ :  $|\Delta_j| \ll \max_i |\mathcal{H}_i^1 \gamma_i^0|$ . Therefore, the values found for  $\gamma_i^0$  are fully suitable for evaluating the sensitivity factors  $T_2$  for the tolerance values of the functionals  $K_{\text{eff}}^{\text{tol}}$ ,  $t_{\text{ce}}^{\text{tol}}$ ,  $t_{\text{ca}}^{\text{tol}}$ ,  $\xi_{\text{ca}}^{\text{tol}}$ ,  $\sigma_w^{\text{tol}}$ , and the permissible controls  $d_{\text{T}}^{\text{min}}$  and  $h^{\text{min}}$ .

Using the correlations (7), (3) and the results shown in Table 1, we obtain

$$\begin{aligned} \frac{\delta T_2}{\delta K_{\text{eff}}^{\text{tol}}} &= +58.28 \text{ years;} \\ \frac{\delta T_2}{\delta t_{\text{ce}}^{\text{tol}}} &= -0.00151 \text{ yr}/^\circ\text{C}; \\ \frac{\delta T_2}{\delta t_{\text{ca}}^{\text{tol}}} &= -0.00810 \text{ yr}/^\circ\text{C}; \\ \frac{\delta T_2}{\delta \xi_{\text{ca}}^{\text{tol}}} &= -12.63 \text{ years;} \end{aligned}$$



$$\frac{\delta T_2}{\delta \sigma_W^{\text{tol}}} = -0.0551 \text{ yr} \cdot \text{mm}^2/\text{kg};$$

$$\frac{\delta T_2}{\delta a_T^{\text{min}}} = +0.198 \text{ yr/mm};$$

$$\frac{\delta T_2}{\delta h^{\text{min}}} = +9.29 \text{ yr}.$$

Emphasizing the importance of evaluating the sensitivity factors  $T_2$  with regard to the examined technological parameters, we will note that the error  $\Delta K_{\text{eff}} = \pm 0.01$  in the value  $K_{\text{eff}}$  produces the same indeterminacy in the value  $T_2$  as the error  $\Delta t_{\text{ce}} = \pm 385^\circ\text{C}$  in the temperature at the center of the fuel element or the error  $\Delta t_{\text{ca}} = \pm 72^\circ\text{C}$  in the temperature of the fuel element casing. These latter errors are comparable to the temperature drops caused by overheating factors ( $370$  and  $80^\circ\text{C}$ , respectively).

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# USE OF INTERNAL DIRECT-CHARGE DETECTORS AS INPUT ELEMENTS OF AUTOMATIC REACTOR CONTROL SYSTEMS

M. G. Mitel'man, L. G. Andreev,  
I. V. Batenin, B. G. Dubovskii,  
V. A. Zagadkin, V. F. Lyubchenko,  
K. N. Mokhnatkin, N. D. Rozenblyum,  
V. S. Sever'yanov, V. B. Tregubov,  
Yu. M. Shipovskikh, and A. I. Shtyfurko

UDC 621.039.562

Automatic power control is one of the basic problems in the operation of nuclear power reactors. Its successful solution depends on the availability of accurate and reliable information of the power and energy release distribution in the reactor core. In principle, a nuclear reactor can be controlled by measuring its power by heat-engineering methods, the  $\gamma$  radiation of the fission products, and the produced neutron flux.

The principal drawbacks of the heat-engineering method is its slow response and limited measuring range, which are respectively determined by the time required for thermal equilibrium to be established during transition periods and by small temperature differences of the coolant at low reactor powers. Because of these difficulties, the heat-engineering method is rarely used for automatic control, but is widely employed as a reference method for calibration of other reactor power control systems and for providing an auxiliary signal for the reactor control system [1, 2].

Power measurement by means of  $\gamma$  detectors is limited by the slow response of such detectors due to the kinetics of  $\gamma$  quanta emission. Emelyanov et al. [3] suggested a method of correcting the response of  $\gamma$  detectors according to which the time dependence of a  $\gamma$ -quanta flux is represented by a sum of exponents with appropriate coefficients. Unfortunately, both the exponents and coefficients depend on the reactor type and on the location and structure of detectors, and must be found separately for each specific case; moreover, even a small change in the detector location necessitates recalculation of these quantities which always involves much computation and experimental work. Because of these reasons the suggested method of improving the response of  $\gamma$  detectors is rarely used in practice.

The neutron method of power monitoring and control [1, 2] is most widely used because it offers the additional advantage that the parameter directly acted upon by control devices is the neutron flux density. The present use of neutron-current ionization chambers located outside the reactor core as input elements for automatic control systems provides no unambiguous relation between the reactor power and detector readings because of the possible "shading" of the detectors by control elements and because of considerable neutron-field gradients in the reactor. Actually, the operation of detectors located outside the core is based on leakage neutrons. In case of a large core, and in particular when the coefficient of reactivity is positive, there is a vital need in core control which is impossible to accomplish with detectors located outside the core.

This makes it very desirable to design automatic reactor control systems based on intracore neutron detectors. Such a method makes it possible to combine the intracore monitoring system of energy release distribution and the automatic reactor control system (which would reduce the total number of neutron detectors and make the system less expensive), to obtain objective and more accurate data on the total reactor power, and to create an intracore automatic control system.

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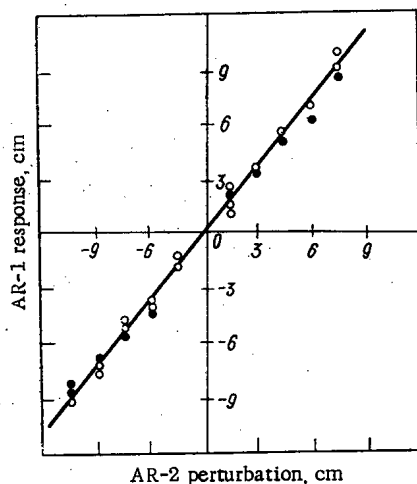


Fig. 1

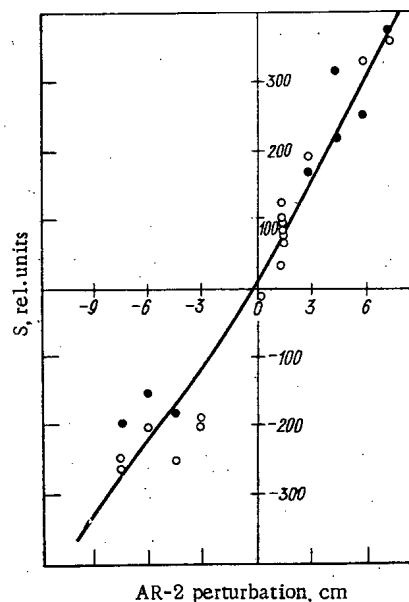


Fig. 2

Fig. 1. Response of AR-1 as a function of perturbation introduced by means of AR-2: ○) direct-charge detectors; ●) ionization chambers.

Fig. 2.  $S$  as a function of perturbation introduced by means of AR-2 (notation the same as in Fig. 1).

Yaremy and Anderson [4] report on the use of direct-charge detectors with a platinum emitter in the automatic control system of a heavy-water reactor. According to [5], such detectors are basically (79%) sensitive to the reactor  $\gamma$  quanta. In addition, 3.4% of its current comes from the activation component with a half-life of 30 min. Thus, automatic control with such detectors has all the disadvantages of  $\gamma$ -detector measurements. The use of direct-charge detectors with platinum emitters for the measurement of energy release distribution in the reactor core is, in addition, associated with difficulties in interpreting the obtained data as they are sensitive to the reactor  $\gamma$  background [6]. Activation direct-charge detectors with rhodium emitters are practically insensitive to reactor  $\gamma$  background.

We have thus investigated the design of a combined system of energy release distribution monitoring and automatic reactor control using activation direct-charge detectors with rhodium emitters. Such detectors are more sensitive than other direct-charge detectors and are preferable for monitoring the neutron flux density (energy release) distribution in the reactor core [7] and, after improving their speed of response with the aid of analog circuits [8], also for automatic reactor control.

An experiment, which confirms the principal possibility of a combined energy release distribution monitoring and reactor control system has been staged on the reactor of the Obninsk Atomic Energy Plant. An energy release monitoring system, consisting of 12 integral DPZ-7 direct-charge detectors uniformly spaced along two mutually perpendicular diameters and two assemblies of four DPZ-1p detectors monitoring the neutron flux density distribution along the core height, has operated at this plant since May, 1972. The instruments measuring the detector currents have the following parameters: current measuring ranges 0 to 10 and 0 to 2  $\mu$ A with a basic error  $\pm 1.5$  and  $\pm 2.5\%$ , respectively; input impedance less than 1 k $\Omega$ ; detector currents recorded with KSP automatic electronic potentiometers. The detector resistance can also be measured.

The amplified detector signals are applied to a summing circuit which also corrects the output signal allowing for the rhodium half-life so that the response of the detector — summing circuit system is practically instantaneous. Signal correction takes into account both decay chains of  $^{104}\text{Rh}$  and  $^{104m}\text{Rh}$  isotopes. Thus, the measuring circuit records the neutron flux density (energy release) distribution in the core, and provides an output signal proportional to reactor power for the automatic control system.

In the course of experiments, we have compared the operation of the automatic reactor control system with external ionization chambers and intracore direct-charge detectors. The reactor power, the position of AR-1 and AR-2 rods of the automatic control system, and the reactivity meter readings were monitored. Perturbations were introduced by changing the position of the AR-2 rods. AR-1 rods were used for con-

trol. Figure 1 indicates that the results of control action obtained with different detectors agree to within  $\pm 8\%$ . Figure 2 shows S as a function of the perturbation of AR-2 rods. The quantity S is equal to the product of the reactor reactivity caused by the perturbation and the time it took to compensate it with AR-1 rods controlled by ionization chambers and direct-charge detectors, and approximately characterizes the response speed of AR-1. Figure 2 indicates that the spread of experimental data obtained with different detectors lies within  $\pm 25\%$ . The total operating time of the reactor power control system with direct-charge detectors was approximately 1 h. No interference effects have been observed. The reactor power remained stable throughout the experiment indicating that control with the aid of intracore direct-charge detectors was quite effective.

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# AN INVESTIGATION OF THE TRANSITION EFFECT IN LAYERED ABSORBERS

V. I. Vit'ko, I. A. Grishaev,  
and G. D. Kovalenko

UDC 539.12.173+539.1.074

The problem of radiation protection has become important with the development of high-energy particle accelerators. High-energy particles passing through a shield generate showers in it. There is an indication in the papers [1-5] that damping of a shower occurs more rapidly in an inhomogeneous medium than in a homogeneous one. In order to design a layered shield, it is necessary to know the quantitative variations of the shower's characteristics in an inhomogeneous medium.

Instruments are employed in high-energy physics whose operation is based on the complete or partial absorption of the initial particle's energy. Such instruments are used mainly to measure the initial particle's energy. Inhomogeneity of the medium spoils the development of a shower, which results in errors in the measurement of the initial particle's energy.

The number of charged particles in the region of a shower's maximum is approximately inversely proportional to the critical energy [6]. Therefore, the number of particles in a shower upon its crossing the boundary separating two media varies inversely as the ratio of their critical energies. The variation of a shower's characteristics upon its passage through inhomogeneous media, which is called the transition effect, is especially noticeable beyond the shower's maximum. A sharp variation in the number of particles results in the fact that it is impossible to neglect the perturbations even for perturbing layer thicknesses of 0.1 radiation length ( $r.l.$ ).

Processes associated with electron absorption due to ionization losses make the largest contribution to the variation in the number of particles in a shower after its passage through a perturbing layer. An indirect measurement of the number of particles in a shower before and after a perturbing layer which would permit estimating the validity of theoretical models for the process in question is of interest.

## Experimental Method

The investigations were conducted on the accelerator of the Kharkov Engineering Physics Institute of the Ukrainian SSR Academy of Sciences. The electron beam's energy was 1 GeV with an energy spread of  $\Delta E/E \sim 1\%$ . The beam current was controlled by a secondary emission monitor. The transition effect was measured after passage of the particles through a copper absorber of variable thickness (0.5, 3, and 8  $r.l.$ ). Behind the main absorber was mounted a perturbing layer of carbon whose thickness was varied from 0.1 to 0.5  $r.l.$  Initially, the number of particles in the shower was measured in the presence of the perturbing layer. Then the perturbing layer was replaced with a copper layer equal to it in radiation length, and the number of particles in the shower was measured.

The number of charged particles in the shower was measured by a secondary emission monitor [7]. The output of electrons from thin foils under the action of fast charged particles can be divided into two groups: the high-energy part — the so-called secondary electrons ejected immediately by the primary particle; and the low energy part — the tertiary electrons ejected from the foil by the secondary electrons.

It was ascertained by Vit'ko et al. [7] that the output of high-energy electrons (secondaries) and the total output of secondary and tertiary electrons depends on the initial particle's energy in the case of thin aluminum foils. The difference between the total output and the secondary electron output, which is equal

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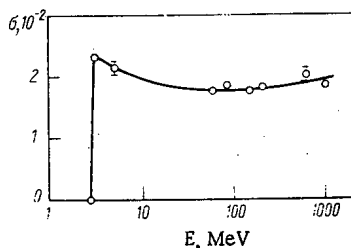


Fig. 1

Fig. 1. Energy dependence of the secondary emission monitor's efficiency.

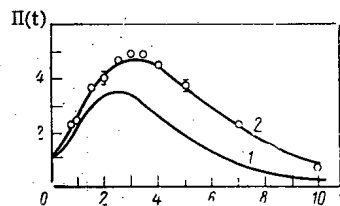


Fig. 2

Fig. 2. Cascade curve for copper: 1 and 2 denote the data from the papers [8] and [9], respectively; ○ denotes the present paper's results.

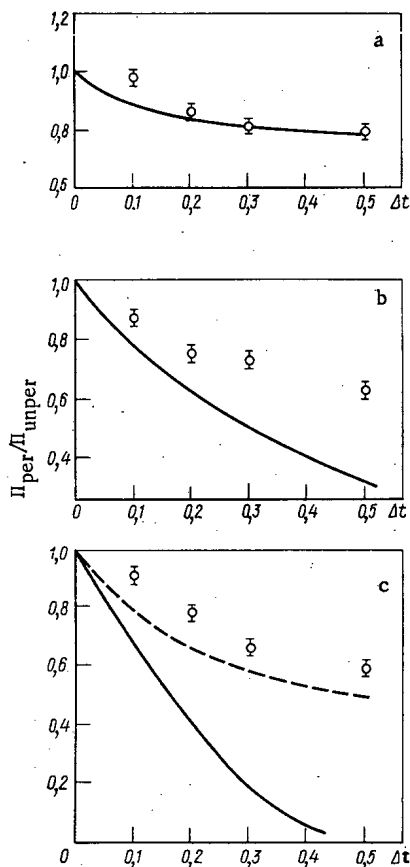


Fig. 3. Dependence of the transition effect on the perturbing layer's thickness for different thicknesses of the principal absorber: — and --- denote the data from the papers [4] and [10], respectively; ○ denotes the results from this investigation.

to the tertiary electron output, in the secondary emission monitor has a weak energy dependence and a sharp drop in the 3–0.15 MeV region.

The dependence of the tertiary electron emission coefficient on the primary particle's energy for the secondary emission monitor is shown in Fig. 1. The sharp decrease in the emission coefficient in the 3–MeV region is explained as being due to the effect of the entry foil. The electrons which arrive from the entry foil in the case of a primary particle with an energy of 3 MeV compensate completely for the departure of electrons from the signal foil. The use of a small energy dependence of the tertiary electron output has permitted reducing the error in the measurement of the number of particles in a shower. The error caused by the emission coefficient's energy dependence is less than 1%.

The total mean-square error in the measurement of the number of particles does not exceed 5% and is caused mainly by the instability of the accelerator's current during the measurements.

### Experimental Results

The experimental results from the measurement of the cascade curve for an energy of 1 GeV in copper which were obtained with the help of a secondary emission monitor having a cutoff threshold of 3 MeV are illustrated in Fig. 2. The absorber's thickness in radiation lengths is plotted along the abscissa, while the number of charged particles in the shower is plotted along the ordinate. Curve 1 represents the theoretical data calculated by the Monte Carlo method with a cut-off threshold of 10 MeV [8], while curve 2 represents the results of the analytic shower theory for a cut-off threshold of 3 MeV [9].

The position of the maximum and the number of particles in the shower agree well with the analytic shower theory. The curve calculated by the Monte Carlo method lies below the experimental points since a cut-off threshold of 10 MeV was used in the calculations, i.e., particles were taken into account which lie in a lower energy region than was measured experimentally.

The agreement of the experimental results with the data from the analytic shower theory [9] confirms that this theory correctly describes the characteristics of a shower in copper for an energy of 1 GeV.

The perturbations introduced by the medium's inhomogeneity depend significantly on the spectrum of the electrons and of the photons. The transition effect has been investigated experimentally behind three

thicknesses of a copper absorber whose choice was due to its isolation of the principal kinds of spectra and our investigation of their effect.

The dependence of the measured ratio of the number of particles in the perturbed shower to the number of particles in the unperturbed shower on the perturbing layer's thickness, in radiation lengths, is shown in Fig. 3. The transition effect after 0.5 (a), 3 (b), and 8 (c) r.l. is illustrated. It is not significant after 0.5 r.l. Beyond such thicknesses the shower spectrum for an energy of 1 GeV contains comparatively more high-energy particles. The low-energy electrons, which after descending from the shower result in a significant decrease in the number of particles, make the principal contribution to the transition effect. There are comparatively few such particles prior to the shower's maximum.

There are more low-energy particles in the spectrum in the region of the shower's maximum (beyond 3 r.l.). On account of this fact, the transition effect is more significant. But the transition effect is especially notable in the shower beyond the maximum (after 8 r.l.). For such a thickness the variation in the number of particles reaches 10% after only 0.1 r.l. and 40% after 0.5 r.l.

The values of the transition effect calculated from the equations of the paper [4] are shown as the solid curves in Fig. 3 for comparison with theory. The variation in the number of electrons in a shower generated in a medium having critical energy  $\varepsilon_0$  at depth  $t$  after the shower's passage through a thickness  $\Delta t$  in a material having critical energy  $\varepsilon$  has been derived by Pinkau [4].

The ratio of the number of particles in the perturbed shower to the number of particles in the unperturbed shower for our case (preserving all the notation used in the paper [4]) is equal to

$$\Pi_{\text{per}}/\Pi_{\text{unper}} = \left\{ \Pi(E_0, 0, t + \Delta t) - \Pi(E_0, \varepsilon_0 \Delta t, t) + \Pi(E_0, \varepsilon \Delta t, t) + 2 \ln \frac{\varepsilon}{\varepsilon_0} \Pi(E_0, 0, t) \frac{4/3 + 2b}{\lambda_1(s) + \mu_0} [1 - \exp(-\mu_0 \Delta t)] \right\} / \Pi(E_0, 0, t + \Delta t). \quad (1)$$

The second, third, and fourth terms in the numerator give the variation in the number of electrons, which is caused by the difference between the materials of the principal absorber and the perturbing layer. The contribution from these three terms is zero for a homogeneous medium ( $\varepsilon = \varepsilon_0$ ). Therefore, they are absent from the denominator. In addition, the terms  $\Pi(E_0, 0, t) + \Delta t \partial \Pi(E_0, 0, t) / \partial t$  in Eq. (1) are combined into a single term:  $\Pi(E_0, 0, t + \Delta t)$ .

Calculations based on Eq. (1) using the spectra of Bhabha and Chakrabarty are shown by the solid curves in Fig. 3. We note that the theoretical data are valid for the region  $\Delta t \geq 0.1$ .

It is evident that this theory [4] provides good agreement with experiment for the high-energy part of the spectrum in a shower generated after 0.5 r.l. of the principal absorber. The discrepancy is significant for the shower's spectrum in the vicinity of the maximum (3 r.l.), and it becomes even greater for the low-energy part of the spectrum. Such a large discrepancy (see Fig. 3) cannot be easily explained by just the presence of a finite cut-off threshold in the experimental apparatus.

A simpler method of treating the transition effect has been adopted by Crannell [10], but it is applicable only to spectra formed far beyond the maximum. For our case we have:

$$\Pi_{\text{per}}/\Pi_{\text{unper}} = \{ \lambda_{\gamma} (\lambda_{\varepsilon_0} - \lambda_{\gamma_0}) \lambda_{\gamma_0}^{-1} (\lambda_{\varepsilon} - \lambda_{\gamma})^{-1} \times [\exp(-\lambda_{\gamma} \Delta t) - \exp(-\lambda_{\varepsilon} \Delta t)] + \exp(-\lambda_{\varepsilon} \Delta t) \} / \exp(-\lambda_{\gamma} \Delta t). \quad (2)$$

Here  $\lambda_{\gamma}$  is the minimum attenuation coefficient for electrons, and  $\lambda$  is the absorption coefficient for electrons. The coefficients with subscript zero refer to copper, and the others refer to carbon.

Calculations based on Eq. (2) are shown in Fig. 3 by the dashed line. They agree better with our data, but the discrepancy is still greater than the experimental errors. Comparing the data from our paper with the theoretical results [4, 10], one can conclude that the experimentally measured transition effect is not as large as one would expect from theory. Models for the transition effect which have been adopted in the papers [4, 10] give only a qualitative description of the experiments and are in need of refinement. Pinkau's theory [4] describes the transition effect well after the maximum, but it is not applicable for a transition layer of 0.1-0.2 r.l.

In conclusion, the authors would like to express their gratitude to the accelerator staff for ensuring the fine operation of the accelerator during these experiments.

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PHASE EXTENSION OF A RELATIVISTIC ELECTRON  
BUNCH INTERACTING WITH A RESONATOR IN A  
STORAGE RING

S. G. Kononenko, N. I. Mocheshnikov,  
and N. N. Naugol'nyi

UDC 621.384.61

In connection with the proposal [1] to use passive resonance elements for additional acceleration of intensive beams of charged particles (autoacceleration), it is of interest to study the influence of this interaction on the equilibrium characteristics of electron bunches in storage rings.

After passage through the resonator, the change in the energy  $\varepsilon(\theta)$  of a particle with azimuthal position  $\theta$  relative to the maximum of the density distribution can be represented sourcewise in terms of  $\rho(\theta)$ :

$$\varepsilon(\theta) = \int_{-\infty}^{\infty} d\xi \rho(\xi) \varphi(\xi - \theta), \quad (1)$$

where the difference kernel  $\varphi(\xi - \theta)$ , the Green's function, is determined by the limiting conditions of the problem.

The redistribution of the energy within the bunch changes the square of the frequency of the incoherent synchrotron oscillations in proportion to the gradient of the variation of the energy of the synchrotron particle:  $\partial_{\theta} = \partial_{\varepsilon}$ . Considering the interaction of a bunch with a cylindrical resonator of length  $h$  and radius  $b$ , we obtain for the normal azimuthal density distribution with variance  $\langle \xi^2 \rangle$  in the ultrarelativistic case to accuracy  $\gamma^{-2}$  ( $\gamma$  is the reduced energy) the following expression from Eq. (1):

$$\partial_{\theta=0} \varepsilon(\theta) = \frac{2N}{\pi^{1/2} h c^2 \langle \xi^2 \rangle^{1/2}} \left\{ 1 - \exp \left( - \frac{h^2}{\langle \xi^2 \rangle} \right) \right\} \sum_{\lambda=1}^{\infty} L(\lambda) \alpha_{\lambda}^{-2}, \quad (2)$$

where  $N$  is the number of particles with charge  $e$ ;  $c$  is the velocity of light;  $L(\lambda)$  is the radial part of the Green's function; and  $\alpha_{\lambda}(b)$  is a root of the Bessel function.

If the bunch length in the case of a low current,  $\langle \xi^2 \rangle_s^{1/2}$ , is determined by the combined influence of quantum fluctuations of the radiation and radiative friction, we find by means of Eq. (2)

$$\frac{\langle \xi^2 \rangle}{\langle \xi^2 \rangle_s} = 1 + L_{\Sigma} \frac{N \langle \xi^2 \rangle^{1/2}}{h \gamma^3} \left\{ 1 - \exp \left( - \frac{h^2}{\langle \xi^2 \rangle} \right) \right\}, \quad (3)$$

where  $L_{\Sigma}$  is a structure constant.

Equation (3) is a generalization of the result of [2] for arbitrary values of the parameter  $h/\langle \xi^2 \rangle^{1/2}$ . The maximal coupling between the resonator and the beam corresponds to the condition  $h \sim \langle \xi^2 \rangle^{1/2}$ . When  $h/\langle \xi^2 \rangle^{1/2} \ll 1$ , we have [2]

$$\frac{\langle \xi^2 \rangle}{\langle \xi^2 \rangle_s} = 1 + L_{\Sigma} \frac{N h}{\gamma^3 \langle \xi^2 \rangle^{1/2}}. \quad (4)$$

The last expression differs qualitatively from the analogous expression in [2] only by the dependence on  $h$ , the flight path. Equation (4) agrees well with the experimental data of [3].

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Original article submitted May 20, 1974

# RELIABILITY OF ELECTRON ACCELERATORS FOR RADIOCHEMICAL DEVICES

Yu. D. Kozlov

UDC 621.384.6:541.15

We have investigated the reliability of the units in electron accelerators [1]. The operation of these machines was monitored over periods of 1.5-3.5 yr. The total operating times of the accelerators, which were in operation during one-shift periods under the rated conditions, were equal to 1000-4000 h.

Table 1 provides data on breakdowns in the units of accelerators of this type with respect to the usual classification characteristics [2]. The statistical data and the equations given in [3] were used for calculating the probability of trouble-free operation  $P(t)$ , the operating time to failure  $T$ , the mean operating time to failure  $t_{me}$ , the failure flow parameter  $\omega(t)$ , and the failure rate  $\lambda(t)$  for the repairable and unrepairable accelerator units. Investigations of the first three accelerators have shown that steady-state operation begins immediately after completion of the start-up and adjustment work, which means that the working-in period has been reduced to two or three months.

The hypotheses concerning the distributions of failures in the units were checked by using the methods of Pearson and Kolmogorov [4, 5]. As a result of analyzing the breakdowns and checking the hypotheses, we found that the operating time to failure of the high-voltage sources and the control panel is characterized by an exponential distribution, while the breakdowns of the cathode unit, the vacuum system, and the outlet window have exponential and normal distributions. Since there were no breakdowns in the tube, the focusing system, the beam scanning system, and the converter unit, the reliability characteristics of these units were calculated by means of the approximate expressions borrowed from [6]. For the number of breakdowns  $m = 0$  [6], we determined the upper and lower confidence limits of the operating time to failure and the failure rate of the above units, i.e.,  $T_u$  and  $T_l$ .

Table 1 provides data on the basic reliability characteristics of the accelerator units, which can be

TABLE 1. Classification of Causes of Failure and Basic Reliability Characteristics of the Units

Unit	No. of breakdowns,* caused by faults					$T, h$	$t_{me}, h$	$T_l, h$	$\lambda \cdot 10^{-3}, h^{-1}$	$\omega \cdot 10^{-4}, h^{-1}$	$\lambda_u \cdot 10^{-3}, h^{-1}$
	structural	technical	operational	wear	sudden failure						
High-voltage source	—	1	—	—	5	1400	—	—	—	7,14	—
Accelerator tube	—	—	—	—	—	—	—	200	—	—	5,0
Cathode unit	—	—	—	4	7	170	—	—	5-9	—	—
Vacuum system with scanning cone	—	—	1	8	11	2000	—	—	—	6,3	—
Beam focusing system	—	1	—	—	—	—	—	170	—	—	5,9
Beam scanning system	—	—	—	—	—	—	—	170	—	—	5,9
Outlet window	—	—	2	17	38	—	30	—	3,3	—	—
Control panel	—	—	—	—	16	1200	—	—	—	8,33	—
Converter unit	—	—	—	—	—	—	—	570	—	—	1,75

\*The breakdowns in units recorded during the working-in period were not taken into account in calculations.

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Abstract submitted May 5, 1975

#### SPECTRAL DISTRIBUTION OF THE ALBEDO OF $^{137}\text{Cs}$ $\gamma$ RADIATION FOR A TWO-LAYER MEDIUM WITH A CYLINDRICAL INTERFACE

Yu. B. Davydov, A. A. Timonov,  
and A. V. Davydov

UDC 550.835

The spectral distribution of the albedo of  $^{137}\text{Cs}$   $\gamma$  radiation is considered for a two-layer medium with a cylindrical interface corresponding to the measurement geometry in a drill hole.

The basic trends in the spectral distribution of the albedo of  $\gamma$  radiation are determined by calculations, supplemented by experimental observations on drill-hole simulators. We have calculated the flux of singly scattered radiation by integrating the  $\gamma$ -radiation fluxes scattered by elementary volumes throughout the scattering medium. The error due to the photoeffect deficit was smaller than 3.5%, while the error due to the deficit of multiply scattered  $\gamma$  quanta did not exceed 25%. The integrals were calculated by using the grid method with the spacings  $r = 0.1$  and  $r = 0.25$  cm. In order to calculate the spectral distribution, the energy range for singly scattered  $\gamma$  quanta was subdivided into 16 intervals. For the chosen method of space and energy subdivision, the calculation error amounted to less than 1%.

We have also investigated the dependence of the flux of singly scattered  $\gamma$  quanta on the radius of the drillhole instrument, the rock density, the drilling-fluid density, the probe length, and the drillhole radius.

The calculation results were checked experimentally on drillhole simulators with a silicate composition. The measurement geometry corresponded to the conditions of theoretical calculations. The detector's resolving power with respect to energy was not less than 15%.

Comparison between the instrumental and the theoretical spectra of  $\gamma$  radiation shows that the theoretical spectra can be used for qualitative interpretation of experimental results. Certain characteristics of the spectral distribution could be of practical interest, for instance, for determining the drillhole diameter and the rock density.

Original article submitted May 27, 1974

Abstract submitted May 6, 1975

# OPTIMIZATION OF THE HEALTH PROTECTION ZONE AND SHIELD PARAMETERS FOR ACCELERATORS

Yu. A. Volchek and A. Ya. Yakovlev

UDC 621.039.58:621.384.6

For accelerators with particle energies of several GeV and beam intensities  $>10^{13}$  particles/sec, the dimensions of the health protection zone must be determined by taking into account the possibility of neutron leakage outside the building and considering the air activated during accelerator operation and discharged into the atmosphere. The dose rate beyond the shield must obey the following relationships (see Fig. 1):

$$D(x, y, h, r) = D_1; \quad (1)$$

$$D(x) = D_2, \quad (2)$$

where  $D_1$  and  $D_2$  are the maximum allowable dose rates at the boundary of the health protection zone and the outside surface of the shield, respectively.

The chosen shield project should provide for minimum cost of the equipment and operation:  $C_{\text{tot}} = C_b + C_s \rightarrow \min$ , where  $C_{\text{tot}}$  is the total cost of protection measures,  $C_b$  is the cost of the building lot, and  $C_s$  is the cost of the shield structure and the ventilation tubes. This can be achieved by varying the parameters on which the cost and efficiency of the shield depend, such as the structural shield thickness, the radius of the health protection zone, and the height of the ventilation tube (see Fig. 1). The problem consists in determining the minimum of the function  $C_{\text{tot}}(x, y, h, r)$  for the conditions (1), (2). By solving the above problem for specific initial functions, we obtain the following relationships:

$$x = \frac{1}{\Sigma_1} \ln \frac{A\beta_1}{D_2 R^2}; \quad (3)$$

$$\frac{r^3 + r^2 \frac{sd + \alpha - b \frac{\Sigma_2'}{\Sigma_2}}{2c_1} + r \left( \frac{D'\beta_3}{s^2 D_1} + \frac{b}{c_1 \Sigma_2} \right) - \left( sd + \alpha - b \frac{\Sigma_2'}{\Sigma_2} \right) \frac{D'\beta_3}{2D_1 c_1 s^2}}{r \frac{R^2 D_2}{D_1} + \frac{R^2 D_2}{2D_1 c_1} \left[ \frac{b}{\Sigma_2} (\Sigma_1' - \Sigma_2') + ds + \alpha \right]} = e^{-\Sigma_1' r}; \quad (4)$$

$$y = \frac{1}{\Sigma_2} \left( \ln \frac{B\beta_2}{D_1 r^2 - \frac{D'\beta_3}{s^2} - R^2 D_2 e^{-\Sigma_1' r}} - \Sigma_2' r \right), \quad (5)$$

where A and B are quantities proportional to the emergence of the leading radiation groups from the side and top shields, respectively,  $D_1$  is a quantity proportional to the concentration of the radioactive air

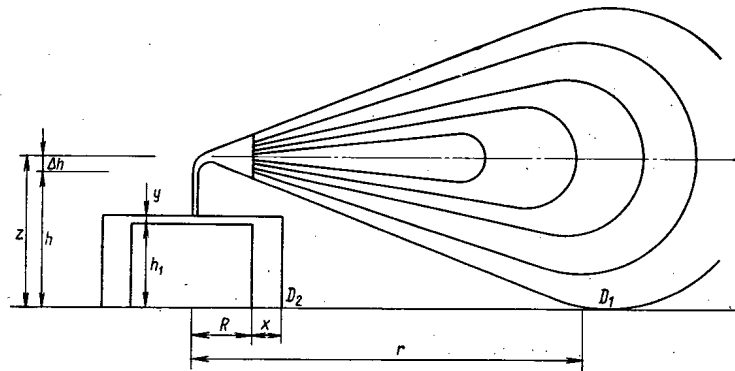


Fig. 1. Diagram for calculating the optimum parameters of the accelerator shield. The symbol R denotes the radius of the accelerator building,  $h_1$  is the building height,  $x$  is the lateral shield thickness,  $y$  is the thickness of the top shield,  $h$  is the height of the ventilation tube,  $\Delta h$  is the elevation from the tube horn to the horizontal axis of the flare,  $z$  is rise height of the radiation refuse to the horizontal axis of the flare, and  $r$  is the distance between the radiation source and the point under consideration.

discharged through the ventilation tube,  $\Sigma_1$  and  $\Sigma_2$  are the effective extraction cross section of the leading groups of direct and scattered radiation in the shield material ( $\text{m}^{-1}$ ),  $\Sigma_1'$  and  $\Sigma_2'$  are the analogous quantities for air,  $\beta_1$  and  $\beta_2$  are the factors of conversion of the radiation flux density into dose rate [ $\text{mrem} \cdot \text{h}^{-1}/(\text{neutrons} \cdot \text{cm}^2 \cdot \text{sec}^{-1})$ ],  $\beta$  is the factor of conversion of the concentration of radioactive isotopes in air into dose rate [ $\text{mR} \cdot \text{m}^3/(\text{h} \cdot \text{mCi})$ ],  $s$  is the turbulence coefficient,  $c_0$  is the land appropriation cost (rubles/ $\text{m}^2$ ),  $c_1 = c_0\pi$ ,  $\alpha$  is the cost of communications per lineal meter of radius of the health protection zone,  $d$  is the cost per meter length of the ventilation tube (rubles/m),  $R$  is the radius of the accelerator building (m), and  $b$  is the cost per meter thickness of the top shield (rubles/m).

The calculations performed for a synchrotron with  $E_0 = 1 \text{ GeV}$  and  $I_0$  values equal to 1, 10,  $10^2$ , and  $10^3 \mu\text{A}$  support the conclusions reached in a paper by Yu. A. Volchek et al., "Design and calculation of buildings and equipment for scientific research" (Nauka, Moscow, 1973), stating that the parameters conforming to the radiation safety standards should be optimized in selecting the construction site and designing the shield, especially when the cost of the land allocated for construction is high. This is of special importance for high-precision equipment, for which the radius of the health protection zone must be greatly enlarged in connection with the discharge of radioactive air into the atmosphere.

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ELEMENTAL ANALYSIS OF BORON CARBIDE AND  
INITIAL COMPONENTS BY PROTON-INDUCED  
X RAYS

A. G. Strashinskii, G. K. Khomyakov,  
N. A. Skakun, N. V. Serykh,  
and I. T. Ostapenko

UDC 539.1.074.55

Elemental analysis of substances through detection of characteristic x rays induced by heavy accelerated particles has evoked great attention among many researchers. In [1-8], the relation of method sensitivity to the nature of the accelerated particles, their energy, and the background intensity was examined; the possibility of quantitative analysis of thick samples was discussed, and a comparison was made with methods employing photon-induced characteristic x rays for analysis.

The present study is devoted to the analysis of boron carbide and its initial components, using characteristic x rays induced by 2.5-MeV protons.

Materials containing boron are widely used in various technological fields. Boron carbide, for example, is used as an absorber in nuclear reactors, as well as in the production of high-temperature semiconducting materials.

Analysis of boron carbide for impurities is a rather difficult task. Spectral methods do not provide the necessary precision, and the analysis of  $B_4C$  from solutions is hampered by the compound's resistance to acids. Melting is usually used for conversion of boron carbide to solution; this results in substantial dilution of samples and, as a result, in lower analysis sensitivity. Moreover, extraneous impurities are introduced during melting, making additional analyses necessary.

TABLE 1. Results of Analysis of Samples Studied

Substance	Impurity content, mass %									
	S	Cl	Ca	Cr	Mn	Fe	Ni	Cu	Zn	Pb
Carbon black	$7,8 \cdot 10^{-1}$	—	$2,8 \cdot 10^{-2}$	—	$1,9 \cdot 10^{-4}$	$6,5 \cdot 10^{-3}$	$3,8 \cdot 10^{-5}$	—	—	—
Boron powder	—	$1,6 \cdot 10^{-1}$	$7,3 \cdot 10^{-3}$	—	—	$6,8 \cdot 10^{-4}$	—	—	—	—
$B_4C$ powder	$3,6 \cdot 10^{-1}$	—	$4,9 \cdot 10^{-3}$	$7,7 \cdot 10^{-4}$	$2,4 \cdot 10^{-4}$	$1,5 \cdot 10^{-2}$	$3,4 \cdot 10^{-3}$	$3,7 \cdot 10^{-4}$	—	—
$B_4C$ plate*	$2,7 \cdot 10^{-1}$	—	$3,3 \cdot 10^{-2}$	$1 \cdot 10^{-3}$	$7,6 \cdot 10^{-4}$	$2,2 \cdot 10^{-2}$	$2,2 \cdot 10^{-3}$	$9,8 \cdot 10^{-4}$	$2,1 \cdot 10^{-4}$	$4,8 \cdot 10^{-2}$
	$\pm 1\%$		$\pm 1\%$	$\pm 9\%$	$\pm 10\%$	$\pm 2\%$	$\pm 7\%$	$\pm 12\%$	$\pm 29\%$	$\pm 3\%$
				$< 1 \cdot 10^{-3}\dagger$	$< 1 \cdot 10^{-4}\dagger$	$1,8 \cdot 10^{-2}\dagger$	$< 1 \cdot 10^{-3}\dagger$	$5 \cdot 10^{-4}\dagger$	$< 5 \cdot 10^{-3}\dagger$	

\*Prepared by a hot molding method.

†Spectroscopic analysis data.

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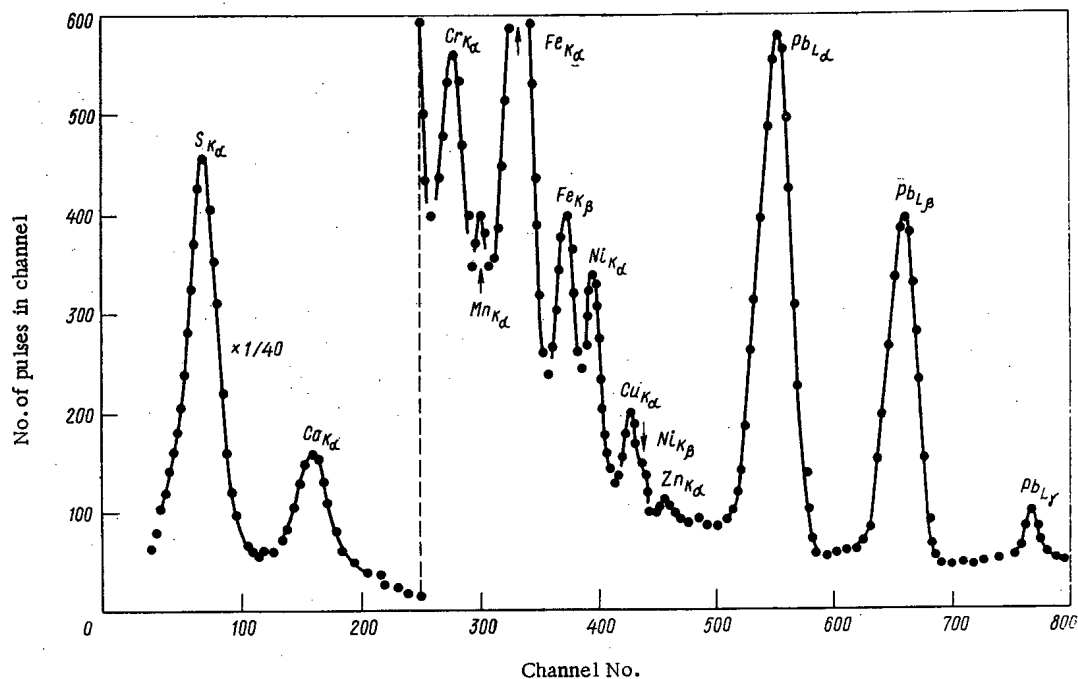


Fig. 1. X-ray spectrum of elements detected in boron carbide.

For the detection of trace elements in boron carbide, it is preferable to use an activation analysis method, or a method using characteristic x radiation induced in samples by accelerated charged particles or photons from various sources. The objects of analysis used were type B-99 amorphous boron, GOST 7885-68 carbon black powder, and plates of boron carbide (see Table 1).

The experimental technique was analogous to that described in an earlier work [9]. Spectrometer energy resolution was 260 eV according to the calibration pulse of a precision amplitude oscillator, and 290 eV for a 6.4-keV line of  $^{57}\text{Co}$ . The size of the proton beam hitting the samples was  $\sim 3 \cdot 10^{-8}$  A. Time of bombardment of each sample was 15 min. The charge of protons incident on a sample was controlled by a surface barrier detector of protons elastically scattered onto a thin carbon film. The detector was positioned in front of the sample to be analyzed. The monitor was periodically calibrated by a charge entering a Faraday cylinder. Powders of boron, carbon black, and boron carbide were placed in packets made from Mylar film 6  $\mu$  thick. The surface of the boron carbide plates was polished on boron carbide powder and washed off with ethyl alcohol. All samples were mounted in an aluminum holder. Background from the holder was practically nonexistent.

It is apparent from Table 1 that, due to processing operations, the boron carbide powder and plates show traces of impurities which do not appear in the initial materials.

To determine relative error, experimental spectra were analyzed by a graphic method allowing for the line form which was dictated by the equipment. The relation of line width to x-ray quantum energy was not taken into account. Since background and signal are statistically independent values, total dispersion of signal and background will amount to

$$\sigma_{s+b}^2 = \sigma_s^2 + \sigma_b^2,$$

where  $\sigma_s$  and  $\sigma_b$  are the standard error in the number of pulses of signal and background, respectively. The confidence interval was taken to be equal to twice the root-mean-square error (a confidence level of 95%).

A standard spectrum of copper applied to mica by vacuum evaporation was used for conversion to mass percents. The amount of copper in the standard was determined by weighing and by proton backscattering [10]. Type ChDA granulated electrolytic copper (analysis grade) was used in preparing the standard.

Figure 1 shows an x-ray spectrum of elements detected in plates of boron carbide. An admixture of lead may appear on the plate surfaces when they are polished on glass. This assumption, however, needs to be checked. It should be noted that the boron powder in this experiment was not analyzed for elements with  $Z > 26$ .

In studying thick samples, the change in the characteristic x-ray production cross section as protons are decelerated, as well as absorption of x rays in the matrix, was taken into account. The number of characteristic radiation quanta which are produced by an impurity element in a thick sample may be estimated for a single proton by the expression

$$n = K \int_{E_0}^0 \sigma_0(E) \frac{e^{-\sigma_1(x\rho)}}{dE/d(x\rho)} dE,$$

where K is the number of atoms of the impurity in 1 g of matrix, g<sup>-1</sup>;  $\sigma_0$  is the emission cross section of the characteristic radiation, cm<sup>2</sup>;  $\sigma_1$  is the coefficient of absorption of characteristic radiation in the matrix, cm<sup>2</sup>/g; E is the initial energy of accelerated protons, MeV; dE/d(xρ) is the stopping power of the matrix material MeV·cm<sup>2</sup>/g; ρ is the density of the matrix, g/cm<sup>3</sup>; x is the range of protons in the matrix up to the production of radiation, and the path traversed by an x-ray quantum in the matrix. This expression was used in processing experimental results on a computer. The description of the procedure for calculating corrections is of interest in itself and will be published in the future. It is advisable here to simply point out that corrections play a significant role in analyzing all elements. If the x-ray production cross section (with reduction in proton energy from 2.5 MeV to 250 keV) decreases on average by a factor of 25 for elements with Z ≈ 15, there is a 600-fold reduction in it for elements with Z ≈ 30. On the other hand, absorption of emitted x rays constitutes only several percent for middle elements, reaching 30-40% and more for the light elements.

When the method of determining trace elements in thick samples is used, the laborious process of preparing thin, homogeneous samples becomes unnecessary, and the absolute quantity of impurity elements may be established without destruction of samples (e.g., in targets for nuclear research, in various monocrystals, etc.). Higher analysis sensitivity is achieved in the determination of trace elements in rather light matrices, which absorb x rays poorly. A negative aspect of thick-layer analysis is the reduction of sensitivity by approximately one order of magnitude in comparison with that achieved in the analysis of thin layers [7].

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# CROSS SECTIONS AND RESONANCE INTEGRALS FOR THE FISSION OF $^{239}\text{Pu}$ , $\text{Am}$ , $\text{Cm}$ , AND $^{249}\text{Cf}$

K. D. Zhuravlev, N. I. Kroshkin,  
and A. P. Chetverikov

UDC 539.172.4

We have used the cadmium difference method to measure thermal-neutron fission cross sections and fission resonance integrals [1, 2] which are of considerable interest for nuclear physics and a number of practical problems such as the optimization of the accumulation of heavy isotopes in reactors.

The measurements were made in a horizontal channel of the SM-2 high-flux reactor using  $582.2 \pm 1.3$  b and  $275 \pm 5$  b [3] for the  $^{235}\text{U}$  thermal-neutron fission cross section and fission resonance integral as reference values. For a cadmium thickness of 1 mm the cadmium ratio for  $^{235}\text{U}$  was  $\sim 40$ . It was assumed that the epithermal neutron spectrum is described by the  $1/E$  law since a water layer  $\sim 40$  mm thick takes part in the formation of the neutron spectrum at the exit from the horizontal channel [4, 5].

Targets of the isotopes under study and the U standard were attached to aluminum substrates 75 mm in diameter and 0.1 mm thick. The weight of material in a target did not exceed  $10 \mu\text{g}$  except for  $^{243}\text{Am}$  for which it was  $\sim 300 \mu\text{g}$ . The number of nuclei in the uranium and plutonium targets was determined by  $\alpha$  counting in  $2\pi$  geometry, using a grid ionization chamber. The number of nuclei in the americium targets was determined by comparing the area of the photopeak of the 59.6-keV  $\gamma$  line with the area of the photopeak of the same line from a standard spectrometric source calibrated with an accuracy of 3%, and by  $\alpha$  counting with a small ( $\sim 10^{-4}$ ) solid angle counter. The number of nuclei determined by the two methods agreed within 2%. The number of nuclei in the curium targets was determined by the number of spontaneous fissions of  $^{244}\text{Cm}$ ,  $^{246}\text{Cm}$ , and  $^{248}\text{Cm}$ . The number of  $^{249}\text{Cf}$  nuclei was determined by  $\alpha$  counting with the small solid angle counter. The number of nuclei in the targets was determined to within 2.5%. The isotopic compositions of the U, Pu, Am, and Cf targets are shown in Table 1; the curium targets had the same isotopic composition as in [6]. The fission fragments were recorded by a double fission chamber. Targets of the isotope being investigated and the standard ( $^{235}\text{U}$ ) were put together back to back and placed in the fission chamber which was located in the neutron beam.

TABLE 1. Isotopic Composition of Targets

Target	MassNo.	Content of mass no., %
$^{235}\text{U}$	244	$1.19 \pm 0.03$
	235	$89.11 \pm 0.18$
	236	$0.61 \pm 0.05$
	238	$9.09 \pm 0.16$
$^{239}\text{Pu}$	239	$99.891 \pm 0.005$
	240	$0.109 \pm 0.005$
$^{241}\text{Am}$	241	100
$^{242m}\text{Am}$	241	$88.97 \pm 0.09$
	242m	$1.03 \pm 0.05$
	243	$10.00 \pm 0.09$
$^{243}\text{Am}$	241	$0.524 \pm 0.019$
	243	$99.476 \pm 0.019$
$^{249}\text{Cf}$	249	100

We have taken account of various experimental errors in the present work. The background of neutrons scattered in the room and from the structural members of the chamber was negligible. No depression in the neutron flux was observed. The following corrections were applied to the results of the measurements: 1) the backscattering of  $\alpha$  particles was taken into account in determining the number of nuclei by  $\alpha$  counting; 2) the isotopic composition of the samples was taken into account; 3) the temperature of the neutrons of the Maxwellian reactor spectrum was estimated as recommended in [4]; 4) the value of  $g(T)$ , taking account of the deviation of the fission cross section from the  $1/v$  law for uranium and plutonium, was taken from [7]; for  $^{241}\text{Am}$  and  $^{242}\text{Am}$  it was determined from the energy dependence of the fission cross section in the thermal region [3, 8],

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TABLE 2. Results of the Measurements

Target	Our data		Data from [5]	
	$\sigma_f$ , b	$I_f$ , b	$\sigma_f$ , b	$I_f$ , b
$^{239}\text{Pu}$	$716 \pm 40$	$328 \pm 22$	$742,5 \pm 3,0$	$301 \pm 10$
$^{241}\text{Am}$	$3,20 \pm 0,15$	$27,7 \pm 1,6$	$3,15 \pm 0,1$	$21 \pm 2$
$^{242m}\text{Am}$	$6080 \pm 500$	$2260 \pm 200$	$6600 \pm 300$	$1570 \pm 110$
$^{243}\text{Am}$	0	$9 \pm 1$	$< 0,072$	$1,5$ [9]
$^{244}\text{Cm}$	$1,0 \pm 0,2$	$13,4 \pm 1,5$	$1,2 \pm 0,1$	$12,5 \pm 2,5$
$^{245}\text{Cm}$	$2070 \pm 150$	$805 \pm 80$	$2020 \pm 40$	$750 \pm 150$
$^{246}\text{Cm}$	$0,14 \pm 0,05$	$13,3 \pm 1,5$	$0,17 \pm 0,10$	$10,0 \pm 0,4$
$^{247}\text{Cm}$	$80 \pm 7$	$730 \pm 70$	$90 \pm 10$	$880 \pm 100$
$^{248}\text{Cm}$	$0,39 \pm 0,07$	$13,1 \pm 1,5$	$0,34 \pm 0,07$	$13,2 \pm 0,8$
$^{249}\text{Cf}$	$1715 \pm 80$	$2200 \pm 100$	$1660 \pm 50$	$21,4 \pm 70$

and for the isotopes of curium and  $^{249}\text{Cf}$  it was assumed equal to unity; 5) the cadmium correction was taken as unity on the basis of measurements made with 0.5 and 1.0 mm cadmium covers. The definitive error of the measurements is determined by the error in finding the number of nuclei, and by the statistical error in measuring the difference in the number of fissions by thermal and resonance neutrons for  $\sigma_f$ , the difference in the number of fissions by resonance neutrons and the number of spontaneous fissions for  $\sigma_f$  (Table 2). In measuring the fission resonance integrals the cutoff energy was fixed at 0.52 eV [1]. For comparison recommended data from [3] are presented and a calculated value of the fission resonance integral of  $^{243}\text{Am}$  from [9]. The good agreement between the values obtained and those recommended for the  $^{239}\text{Pu}$  cross sections shows the reliability of our measurements.

In conclusion the authors thank Yu. S. Zamyatnin and V. N. Nefedov for constant interest in the work and helpful discussions, V. N. Polynov for supplying the curium samples, and V. G. Polyukhov for analyzing the  $\alpha$  spectra.

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# INSTRUMENTAL NEUTRON ACTIVATION ANALYSIS OF GEOLOGICAL AND BIOLOGICAL OBJECTS USING A COMPUTER

V. B. Zlokazov, L. P. Kul'kina,  
and O. D. Maslov

UDC 543.53:681.142.4

The interpretation of  $\gamma$ -ray spectra is the most time-consuming task in activation analysis of complex multicomponent objects. Extraction of all their information is impossible without the use of a computer.

A principle is put forward in this paper for the organization of a computer program applicable to the analysis of  $\gamma$ -ray spectra obtained with the help of any amplitude analyzer, easily controlled, and easily able to be corrected. A search for  $\gamma$ -ray lines which have not been marked in advance is conducted with the help of a library of isotopes.

The program was set up for instrumental neutron activation analysis when several elements are to be determined simultaneously in samples of different origin (geological, biological).

Samples of mummies and lichen were selected for analysis (see Table 1). Sample specimens weighing 0.155 and 0.63 g, respectively, were placed into aluminum cassettes. The irradiation was carried out in a reactor with a thermal neutron flux of  $5 \cdot 10^6$  neutrons/cm<sup>2</sup>.

TABLE 1. Results of the Instrumental Neutron Activation Analysis of Mummy and Lichen Samples

Element	Radioisotope	Content, %	
		mummies	lichen
Sodium	<sup>24</sup> Na	1,0	$5,2 \cdot 10^{-3}$
Scandium	<sup>46</sup> Sc	$5,6 \cdot 10^{-6}$	$2,0 \cdot 10^{-6}$
Calcium	<sup>47</sup> Ca	2,3	—
Chromium	<sup>51</sup> Cr	$6,0 \cdot 10^{-4}$	$2,4 \cdot 10^{-5}$
Iron	<sup>59</sup> Fe	0,14	$5,0 \cdot 10^{-3}$
Cobalt	<sup>60</sup> Co	$1,3 \cdot 10^{-3}$	$1,7 \cdot 10^{-6}$
Copper	<sup>64</sup> Cu	$6,5 \cdot 10^{-4}$	—
Zinc	<sup>65</sup> Zn	$3,7 \cdot 10^{-3}$	$9,0 \cdot 10^{-4}$
Selenium	<sup>75</sup> Se	$5,3 \cdot 10^{-5}$	$< 10^{-5}$
Arsenic	<sup>76</sup> As	—	$< 10^{-5}$
Bromine	<sup>82</sup> Br	$2,4 \cdot 10^{-3}$	$6,0 \cdot 10^{-5}$
Strontium	<sup>85</sup> Sr	0,1	—
Rubidium	<sup>86</sup> Rb	$1,0 \cdot 10^{-3}$	$7,4 \cdot 10^{-5}$
Molybdenum	<sup>99</sup> Mo	$1,3 \cdot 10^{-3}$	—
Silver	<sup>110</sup> Ag	$3,9 \cdot 10^{-4}$	$< 10^{-7}$
Cadmium	<sup>115</sup> Cd	$2,5 \cdot 10^{-3}$	—
Tin	<sup>125</sup> Sn	0,96	—
Antimony	<sup>122</sup> Sb, <sup>124</sup> Sb	$1,3 \cdot 10^{-2}$	$1,3 \cdot 10^{-5}$
Cesium	<sup>134</sup> Cs	$1,7 \cdot 10^{-5}$	$7,0 \cdot 10^{-6}$
Lanthanum	<sup>140</sup> La	—	$2,2 \cdot 10^{-5}$
Cerium	<sup>141</sup> Ce	—	$8,5 \cdot 10^{-6}$
Europium	<sup>152</sup> Eu	$1,2 \cdot 10^{-6}$	$1,5 \cdot 10^{-7}$
Ytterbium	<sup>169</sup> Yb	—	$< 5,4 \cdot 10^{-7}$
Hafnium	<sup>181</sup> Hf	$1,6 \cdot 10^{-6}$	$4,0 \cdot 10^{-6}$
Gold	<sup>198</sup> Au	$1,4 \cdot 10^{-5}$	$< 10^{-7}$
Mercury	<sup>203</sup> Hg	$6,4 \cdot 10^{-5}$	$< 5 \cdot 10^{-6}$

The  $\gamma$ -ray spectra of the irradiated samples, which were exposed for 2-40 days, were measured by a Ge(Li) detector having a volume of 35 cm<sup>3</sup> at a resolution of 3.0 keV for the <sup>137</sup>Cs  $\gamma$ -ray line with  $E = 0.662$  MeV on an AI-4096 amplitude analyzer and a TRA-1001 minicomputer with punched-tape output. The TRA-1001 permits carrying out a preliminary marking out and simple analysis of the  $\gamma$ -ray spectra with the aid of the JUPITER-16K program [1]. Finally, the spectrum was processed on a Minsk-32 computer according to the procedure which we worked out. The  $\gamma$ -ray spectrum of the mummies, which was drawn by a graphical plotter from a punched tape obtained from the computer, is presented in Fig. 1.

The processing on the Minsk-32 computer was carried out using SPORS (spectrum orientation system) [2]. The basis of SPORS is a library of programs, notably a special set of programs in each of the Minsk-32 computer's input languages consisting of standard programs. The library's structure is modular. The class of processing algorithms has been broken into simple parts, and each is realized in a separate program. The replacement of any component is carried out by simply removing or adding the appropriate program.

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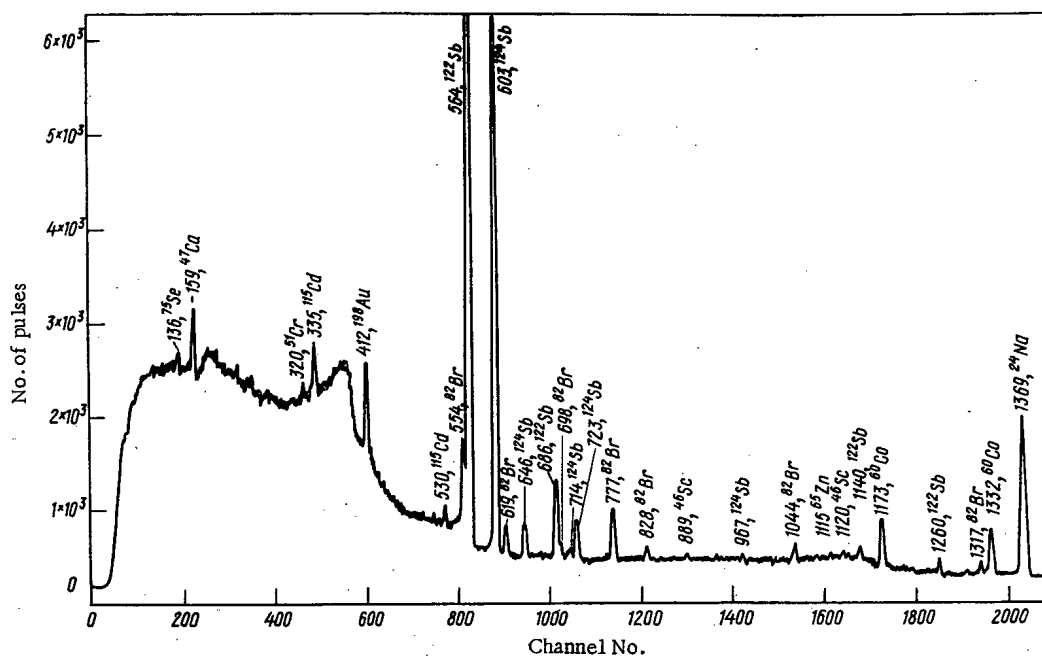


Fig. 1.  $\gamma$ -Ray spectrum of mummies obtained after 9 days of cooling.

The  $\gamma$ -ray spectra obtained with the help of any amplitude pulse analyzer are punched out on paper tape or they are placed into the computer's memory via a communication channel. Each  $\gamma$ -ray spectrum is represented by a set of numerical data treated as a sample trajectory of some random noncoherent Gaussian process. Smoothing of the  $\gamma$ -ray spectra is done by the method of least squares. Processing of the  $\gamma$ -ray spectra includes three stages: adjustment of the spectra taking account of peculiarities in the spectrometer circuit (dead time for encoding, detector efficiency); the mathematical processing (finding the center of gravity of a peak and its area); and identification of the photopeaks and calculation of the content of elements based on a formula utilizing a library of isotopes.

A calibration spectrum of  $^{266}\text{Ra}$  is used to derive the dependences of resolution on energy and energy on the channel. The calibration curve in both cases is taken in the form of a polynomial of the first or second degree.

The library of isotopes, which consists of two sections (a block of isotope descriptions and a block of energy descriptions and their quantum efficiencies for each isotope), is used to search for all the  $\gamma$ -ray lines of all the individual isotopes in the spectrum, which permits assessing overlaps. In case of an overlap of  $\gamma$ -lines, their mathematical separation is provided for by virtue of additional conditions introducible into the assignment of a given section of the spectrum for processing.

Upon completion of the processing of a spectrum, symbols are printed out showing the identified isotopes, the energy of the photopeaks from which these isotopes were identified, the content (in grams per gram) of the corresponding elements in the sample, and the accuracy of the content determination.

In conclusion, the authors thank G. N. Flerov and V. S. Barashenkov for their constant interest in this work and their support, V. Ya. Voropaev for discussion of the results, and B. V. Fefilov and L. P. Chelnokov for providing for the precise operation of the spectrometer equipment and the computer.

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# DETERMINATION OF MANGANESE IN ALUMINUM BY NEUTRON ACTIVATION WITH A $^{252}\text{Cf}$ SOURCE

K. Sailer, Sh. Darotsi,  
Sh. Nad', P. Raich,  
I. Chikai, and L. Gergei

UDC 543.53

In instrumental activation analysis, increasing use has recently been made of  $^{252}\text{Cf}$  spontaneous-fission neutron sources. Such a source has a neutron yield which is high and practically constant and is easy to work with. At the same time a  $^{252}\text{Cf}$  source is cheap enough for industrial laboratories where problems are often encountered which can be solved by means of a source containing about 1 mg of  $^{252}\text{Cf}$ .

Owing to the large cross section of the  $(n, \gamma)$  reaction for many elements, activation by thermal neutrons is preferable if the matrix does not strongly absorb slow neutrons. For example, aluminum is such a matrix, and therefore some components and impurities in aluminum alloys can be determined with high precision with the aid of a simple  $^{252}\text{Cf}$  isotope source. Among the principal components of aluminum alloys, manganese has the greatest cross section for the  $(n, \gamma)$  reaction for thermal neutrons:  $\sigma_{\gamma}(^{55}\text{Mn}) = 13.3 \pm 0.2 \text{ b}$  [1]. Therefore, manganese is the most suitable element for testing the possibility of using  $^{252}\text{Cf}$ .

The aim of our present work was to estimate the accuracy, sensitivity, necessary irradiation and measurement times, etc., for the quantitative analysis of manganese in aluminum with a  $660 \mu\text{g } ^{252}\text{Cf}$  source, a  $40 \text{ cm}^3 \text{ Ge(Li)}$  detector (resolution 3.2 keV at 661.6 keV), and a Didak-4000 analyzer. The absolute efficiency of the  $\text{Ge(Li)}$  detector at 50-1400 keV was determined by means of calibrated  $^{226}\text{Ra}$  and  $^{182}\text{Ta}$  sources of the same dimensions as the specimen [2]. To moderate the fast neutrons we used a water tank 60 cm in radius and 80 cm deep.

To find the optimum irradiation site we investigated the fluxes of thermal, resonance, and fast neutrons in relation to the radial distance  $r$  and vertical distance  $Z$  from the source (Fig. 1). The fluxes of thermal and resonance neutrons were measured by means of Rh, Au, and In foils 63.0, 0.85, and 0.30-4.71  $\text{mg/cm}^2$  thick respectively. The integral fluxes of fast neutrons were measured by means of the threshold reactions  $^{115}\text{In}(n, n')$ ,  $^{27}\text{Al}(n, p)$ , and  $^{27}\text{Al}(n, \alpha)$ ; the thicknesses of the indium and aluminum disks were, respectively, 290 and 620  $\text{mg/cm}^2$ . The diameters of the specimens were the same as those of the AlMn alloy specimens. From the measured values of the fluxes of thermal and fast neutrons we obtained an approximate optimum for the irradiation in the range of distances 5-7 cm from the surface of the source box.

TABLE 1. Quantity of Manganese in AlMn Alloys, Determined by Various Methods,  $10^{-6}$

Specimen No.	Activation method	Spectral absorption method	Method of resistivity measurement
1	$96.8 \pm 2.5$	100	—
2	$297 \pm 7$	300	—
3	$3070 \pm 70$	3000	—
4	$19500 \pm 450$	18700	—
5	$103 \pm 3$	—	105
6	$518 \pm 12$	—	520
7	$676 \pm 19$	—	—
8	$788 \pm 21$	—	—
9	$2850 \pm 70$	—	2840
10	$6420 \pm 150$	—	6170
11	$15800 \pm 360$	—	5270
12	$30800 \pm 780$	—	—

The AlMn alloys were of high purity with  $(100-30,000) \cdot 10^{-6}$  manganese. From these we machined specimens 11 mm in diameter and 3 mm thick with a total mass of 850 mg. Irradiation was carried out at  $Z = 0$  and  $r = 5 \text{ cm}$ ; the irradiation and measurement times were varied from 0.5 to 5 h, and the cooling time was 50 min (to avoid superimposition of the 846.6 and 843.76 keV  $\gamma$  lines [3] of the isotopes  $^{56}\text{Mn}$  and  $^{27}\text{Mg}$ , respectively). The absolute activities of the AlMn specimens and the gold foils used to determine the absolute flux of thermal neutrons were measured with a  $\text{Ge(Li)}$  detector. The flux of thermal neutrons at the irradiation

L. Kosuth University, Debrecen, Hungary. Translated from Atomnaya Énergia, Vol. 39, No. 4, pp. 288-289, October, 1975. Original article submitted February 6, 1975.

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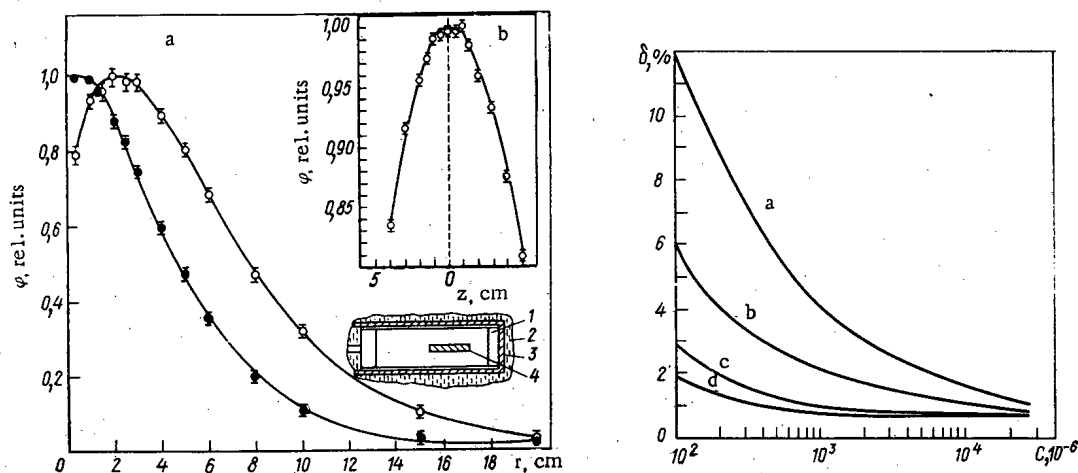


Fig. 1. Neutron fluxes vs distance to surface for  $Z = 0$  (a) and vs depth at  $r = 5$  cm (b). ○) Thermal neutrons; ●) resonance neutrons; 1) source box; 2) water; 3) Plexiglas tube; 4)  $\text{Cf}_2\text{O}_3$ .

Fig. 2. Relative error of analysis based on standards vs time and manganese concentration.  $T_{\text{cool}} = 50$  min;  $T_{\text{irr}} = T_{\text{meas}} = 0.5$  (a), 1 (b), 2.5 (c), or 5 h (d).

TABLE 2. Sensitivity of Determination of Manganese,  $10^{-6}$

Irradiation geometry, $r$ , cm ( $Z = 0$ )	$L_Q^{10\%}$	$L_Q^{33\%}$	$L_D^{95\%}$
0,3	9,7	2,7	3,0
2	5,1	1,4	1,5
5	4,1	1,0	1,1
7	5,3	1,3	1,4

site at  $r = 5$  was  $\Phi_{\text{th}} = (5.64 \pm 0.07) \cdot 10^6 \text{ cm}^{-2} \cdot \text{sec}^{-1}$ . We made corrections for self-screening of the specimens, disturbances of the flux, superimposition of resonance neutrons, self-absorption of  $\gamma$  quanta, and pulse pile-up.

Table 1 compares the manganese contents found by the neutron activation method with the values found by other methods. The activation determinations agree with the results of the other methods. The discrepancy in specimen No. 11 can be explained by the fact that the

resistivity measurement method is suitable only at low manganese concentrations for which the alloy is a solid solution.

From the measurement results we determined the sensitivity of the quantitative analysis with relative standard deviations of 10% and 33% and the sensitivity of qualitative analysis at the 95% confidence level in relation to the experimental geometry (Table 2), as calculated by the Currie method [4]; the data in Table 2 were obtained with  $T_{\text{irr}} = T_{\text{meas}} = 5$  h and  $T_{\text{cool}} = 50$  min.

If the analysis is made with the aid of several standard AlMn specimens, it is interesting to estimate the relative error of the analysis in relation to the time and concentration. The results of these calculations are plotted in Fig. 2; in the calculations referring to the geometry  $Z = 0$ ,  $r = 5$  cm, we neglected the standardization error. From Fig. 2 we see that, even with very short times (about 2 h), large concentrations of manganese can be determined to within better than 5%. The cooling time can be reduced to a few minutes and the analysis time simultaneously reduced to 1 h. In this case, of course, it is necessary to take account of the contribution made by the  $\gamma$  line of  $^{27}\text{Mg}$  at 843.76 keV.

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# SOME RESULTS OF AN EXAMINATION OF A DISMANTLED RK-L HOT LOOP

D. M. Zakharov, V. V. Gavar,  
A. S. Dindun, and M. M. Kramer

UDC 621.039.8:621.039.574

In constructing hot loops for industrial purposes, it is important to study  $\gamma$  carriers and structural materials of hot loops after prolonged use in hot conditions and subsequent dismantling.

In this article we discuss some results of a metallurgical investigation of parts of the communications of a dismantled hot loop of type RK-L (IF AN LatvSSR) after 20,000 h operation under radiation. The design and operating conditions of the RK-L hot loop are similar to those described in [1, 2]. The communications ducts of the loop were made of steel 1Kh18N9T; the  $\gamma$  carrier was a ternary alloy (25 wt. % indium, 23 wt. % tin, rest gallium); the temperatures of various parts of the installation ranged from 15 to 170°C; and the ratio of the volume of  $\gamma$  carrier to the internal area of the loop was about 0.1 cm.

A year after the RK-L hot loop was dismantled, the activity of its spherical irradiator, the parts of the pump channel, and the welded joints of the flowmeter and ducts was negligible; this facilitated investigation of the surfaces of these specimens. The internal surfaces of all the parts of the loop were first treated with alcohol, 8% NaOH solution at 70–80°C, and alcohol again. It is known [3] that this treatment of the surface removes alloys coating the communications parts. Preservation of alloy-amalgamated parts should indicate the development of wetting, i. e., the initial stage of interaction between the structural material and the liquid metal. Careful inspection of the treated surface of the specimens under the microscope revealed local regions of wetting.

These regions were absent only in the surface of the spherical irradiator. No intermetallic compounds, with a number of specific features [3–5], formed by interaction of stainless steel with indium–gallium or indium–gallium–tin alloys, were found on any of the test specimens. After inspection the specimens were subjected to activation analysis, \* but first they were cut up so as to separate the wetted regions from those not wetted by the alloy. The activation analysis was carried out in an AI-100 analyzer with an NaI(Tl) crystal after irradiation of the specimens with neutrons in a nuclear reactor. The analysis revealed that the regions wetted by the alloy contained gallium and indium. In the regions not wetted by the alloy these components were not found. The identification was made by means of the products of the following reactions:  $^{71}\text{Ga} + n \rightarrow ^{72}\text{Ga} + \gamma$  ( $E = 840$  keV) and  $^{115}\text{In} + n \rightarrow ^{115\text{m}}\text{In} + n'$  ( $E_{\gamma} = 335$  keV).

The presence of tin in the regions wetted by the alloy was not observed, because it is difficult to identify tin owing to the interfering activities of the components of steel 1Kh18N9T. The quantitative contents of gallium and indium in the regions wetted by the alloy were determined graphically by comparison with standards. According to the averaged data, the gallium content was 50 times less than the indium content ( $2 \cdot 10^{-2}$  and  $4 \cdot 10^{-4}$  g/cm<sup>2</sup>, respectively). Gallium plays the leading part in the formation of regions of wetting on the surface of the steel.

The data on the interaction between the structural material of the RK-L plant and the indium–gallium–tin alloy disagree with those in [3–5]. The temperature of most of the units and communications in the RK-L hot loop did not exceed 100°C, and at such a low temperature steel 1Kh18N9T should not be wetted

\*The activation analysis was carried out at the Institute of Physics, Academy of Sciences of the Georgian SSR.

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by the alloy. We must emphasize that the radiation did not influence the development of the corrosion process because none of the units and communications of the RK-L was in the zone of intense neutron irradiation. It is true that the integral dose of gamma rays in the irradiator was  $3 \cdot 10^{10}$  rad, but the internal surface of the irradiator was not itself wetted by the alloy.

We must bear in mind that the internal surfaces of the units and communications of the RK-L (except for the surface of the spherical irradiator which was only treated with alcohol to remove contaminants) were subjected to etching and passivation. The etchant was 10 or 20% HCl or  $H_2SO_4$ ; etching was performed at 80-90°C for 2-3 h. Subsequent passivation of the surface (after thorough washing with hot water under pressure) was by treating with 30%  $HNO_3$  solution at room temperature for 2-4 h. In factory conditions steel 1Kh18N9T is usually passivated with 4-7%  $HNO_3$  solution at 70-80°C, which forms an oxide film on its surface with good adhesion and strength properties. Etching the surface led to solution of this oxide film; the composition of the etchant and the etching conditions were not altogether well chosen: In hydrochloric acid at 80-90°C, steel 1Kh18N9T is corroded [6], which is very undesirable. The material was passivated with very concentrated nitric acid but without heating. In this case the material may have been underpassivated or overpassivated. In either case the oxide film does not acquire the required protective properties, which are composed of a whole series of interrelated components: presence of oxides of trivalent chromium in surface layer, optimum values of film thickness, optimum strength, density, and adhesion.

Thus interaction of steel 1Kh18N9T with indium - gallium - tin alloy at low temperature, as in the RK-L, was the result of insufficiently careful chemical treatment of the structural material. In the RK-L irradiator this interaction did not occur, because its internal surface was not etched or passivated and retained its original oxide film with adequate protective properties. The results show that steel 1Kh18N9T used as a structural material for hot loops (up to  $t \leq 100^\circ C$ ) does not need special chemical treatment.

We must emphasize that the appearance of regions of wetting on the surfaces of the units and communications of a hot loop is an undesirable phenomenon. It did not adversely affect the operation of the RK-L loop, but in industrial plants with longer communications, greater volumes of  $\gamma$  carriers, and higher temperatures, the appearance of regions of wetting might reduce the strength of the structural material owing to adsorption. It is known that indium is a surface-active substance with respect to steel at temperatures above 180°C [7].

Attention is also merited by another phenomenon displayed during disassembly of the RK-L hot loop: formation of deposits in the form of a viscous pasty mass on the internal surfaces of the units and communications. If the ternary alloy can be extruded from these deposits in a press, what is left is a black, finely dispersed powder, which, according to a chemical analysis made at the Institute of Chemistry of the Ural Science Center, contains 21.6 mass % Ga, 51.8 mass % In, and 9.9 mass % Sn. The remainder (16.7 mass %) is mostly oxygen. An attempt to make an x-ray structural analysis of the powder was unsuccessful; no reflections could be seen in the diffraction patterns. This is apparently because of the high dispersity of the powder and its content of amorphous substances, since it is known [8, 9] that some oxides of indium, gallium, and tin are amorphous. The oxides may have been in the original metals or may have been formed in the circulating  $\gamma$  carrier. The separated black powder is easily wetted by the alloy, forming viscous pasty deposits which might block the hot loop. Therefore in building the plant we must increase the stringency of the requirements for purity of the original metals, for the treatment of the  $\gamma$  carrier and its filling of the loop, for the cleanness of the interior surface of the communications and units of the plant, and for its vacuum tightness.

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RATIO OF RADIATIVE CAPTURE AND FISSION  
CROSS SECTIONS FOR  $^{239}\text{Pu}$  AT NEUTRON  
ENERGIES OF 0.2-30 keV

A. A. Bergman, A. N. Medvedev,  
A. E. Samsonov, and V. A. Tolstikov

UDC 539.125.5

The method of spectrometric measurement based on retardation times in lead (SRT) has a number of features discussed in [1-3]. The essential features of the method are as follows.

1. Scattering of neutrons by the specimen does not alter the isotropic neutron field of the spectrometer, and hence the detector background is not altered. To test the influence of moderation in the specimen on the background count we made measurements with large graphite specimens. We found that moderation in specimens with plutonium dioxide has a negligible influence on the background count in the measurement conditions.

2. Capture and fission events in the specimen are registered by a proportional  $\gamma$  counter with an efficiency which is proportional to the total energy of the  $\gamma$  quanta emitted during capture and fission, and which is independent of the form of the spectrum of  $\gamma$  quanta.

3. Normalization is carried out in the graphite prism with a well-thermalized isotropic spectrum of neutrons, and this enables us to use the most reliable reference data. Screening for thermal neutrons in the specimens used is small and can be exactly allowed for.

These features enabled us to make methodologically independent measurements of the parameter  $\alpha$  for  $^{239}\text{Pu}$  by SRT. The method of measurement and the preliminary results are given in [1-3].

Table 1. Results of Measurements of  $\alpha$  and  $\sigma_f$ .

E, keV	$\bar{\alpha}$	$\Delta\bar{\alpha}^*$	$\Delta\bar{\alpha}^\dagger$	$\bar{\sigma}_f$	$\Delta\bar{\sigma}_f^*$	$\Delta\bar{\sigma}_f^\dagger$
25-30	0,45	0,02	0,05	1,77	0,03	0,03
20-25	0,51	0,03	0,05	1,79	0,03	0,04
15-20	0,55	0,02	0,05	1,81	0,02	0,04
10-15	0,61	0,02	0,06	1,98	0,02	0,04
9-10	0,74	0,04	0,06	2,01	0,04	0,04
8-9	0,76	0,04	0,06	2,09	0,05	0,05
7-8	0,81	0,04	0,07	2,11	0,05	0,04
6-7	0,86	0,03	0,07	2,21	0,04	0,04
5-6	0,78	0,05	0,07	2,42	0,04	0,05
4-5	0,80	0,04	0,07	2,61	0,06	0,05
3-4	0,84	0,04	0,07	2,92	0,04	0,06
2-3	0,98	0,02	0,08	3,34	0,03	0,07
1-2	1,02	0,02	0,08	4,92	0,04	0,10
1-0,9	0,90	0,05	0,08	6,41	0,14	0,13
0,8-0,9	0,88	0,04	0,08	6,29	0,16	0,13
0,7-0,8	0,98	0,04	0,08	6,37	0,11	0,13
0,6-0,7	1,15	0,03	0,09	7,29	0,10	0,15
0,5-0,6	0,95	0,03	0,08	9,63	0,11	0,19
0,4-0,5	0,76	0,02	0,06	10,4	0,11	0,21
0,3-0,4	0,95	0,03	0,08	12,1	0,11	0,24
0,2-0,3	0,74	0,01	0,06	17,0	0,10	0,32

\*Rms error.

†Error due to normalization.

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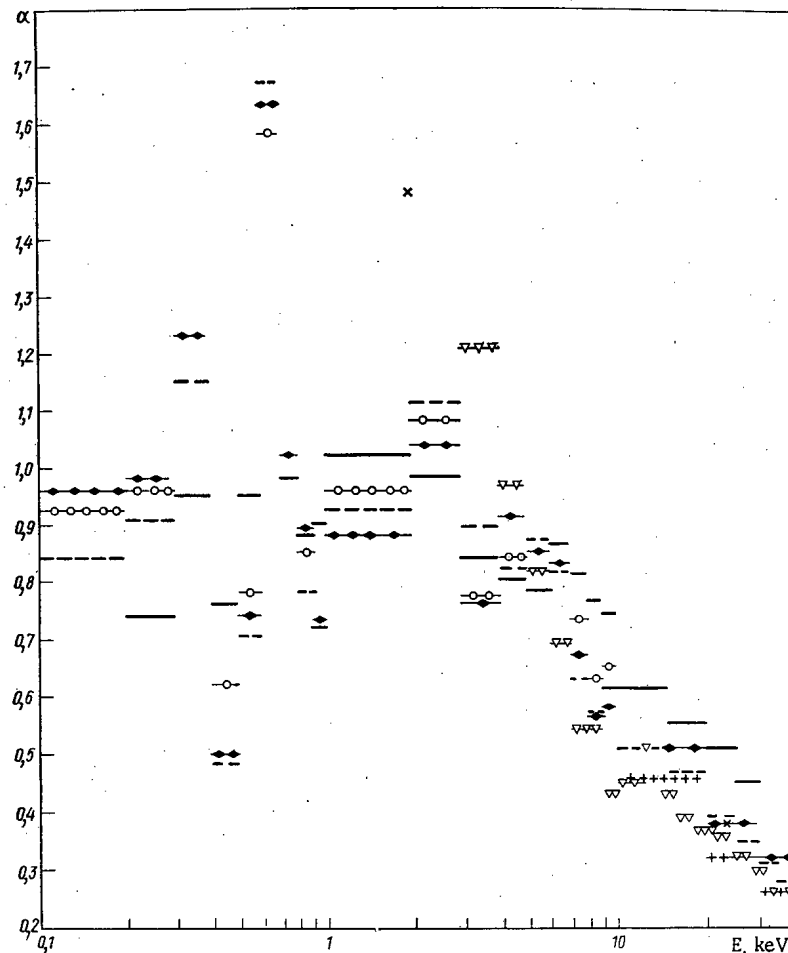


Fig. 1. Averaged values of  $\alpha$  for  $^{239}\text{Pu}$  in comparison with other authors' data ( $\alpha = \sigma_c/\sigma_f$ ). Results: —) present authors; +) [4];  $\nabla$ ) [5];  $\odot$ ) [6];  $\times$ ) [7];  $\blacklozenge$ ) [8]; ---) [9].

In our present work we improved the stability of the electronics. We made more careful measurements of the delay time of registration of fission events in the fission chamber. In the measurements with the fission chamber we reduced the duration of the neutron burst to  $0.5 \mu\text{sec}$ , equal to the duration of the burst for measurements with a  $\gamma$  counter. The new fission data reduced the value of  $\alpha$  in the range  $E_n > 8 \text{ keV}$ .

In our measurements, as well as a "thin" specimen of plutonium with  $\bar{n} = 4.2 \cdot 10^{20} \text{ nuclei/cm}^2$  we used an additional "thick" specimen with  $\bar{n} = 4.9 \cdot 10^{21} \text{ nuclei/cm}^2$ . At  $E_n > 6 \text{ keV}$  where the influence of resonance screening is small, the ratio of the count rates of the  $\gamma$  counter should be constant; this was confirmed by the experiment, and so by using the data from the "thick" specimen we could reduce the statistical scatter in this energy range.

Normalization of the data with respect to  $\alpha$  was effected with thermalized neutrons in a graphite prism with a gold specimen with  $\bar{n} = 1.8 \cdot 10^{21} \text{ nuclei/cm}^2$  and with a "thin" plutonium specimen for which screening in the thermal flux was small.

The measurements with the "thick" specimen were related to the measurements with the "thin" specimen in the energy range  $8 \text{ keV} < E_n < 23 \text{ keV}$ .

The values of  $\alpha(E)$  were calculated from the expression

$$\alpha(E) - \alpha(T) = \left[ \frac{I_{\gamma}^{\text{Pu}}(E) \Delta I_f^{\text{Pu}}(T)}{I_f^{\text{Pu}}(E) \Delta I_{\gamma}^{\text{Pu}}(T)} - 1 \right] \left[ \frac{\Delta I_{\gamma}^{\text{Pu}}(T) \sigma_c^{\text{Au}}(T) P_{\text{Au}} A_{\text{Pu}} B_{\text{Au}}}{\Delta I_{\gamma}^{\text{Au}}(T) \sigma_f^{\text{Pu}}(T) P_{\text{Pu}} A_{\text{Pu}} B_{\text{Pu}} \delta} \right],$$

where  $\alpha(T) = 0.398 \pm 0.004$  is the value of  $\alpha$  for a Maxwellian neutron spectrum at  $22^\circ\text{C}$ ;  $\Delta I_{\gamma}^{\text{Pu}}(T)$  and  $\Delta I_{\gamma}^{\text{Au}}$  are the numbers of counts of the  $\gamma$  counter due to capture of thermalized neutrons in specimens of Pu and Au in the graphite prism;  $\Delta I_f(T)$  is the number of counts of the fission chamber in the graphite

prism;  $I_f^{Pu}(E)$  and  $I_\gamma^{Pu}(E)$  are the numbers of counts of the fission chamber and  $\gamma$  counter (due to capture and fission in the  $^{239}Pu$  specimen) at the moment of time of the cycle of the corresponding average neutron energy  $\bar{E}_n$ ;  $\sigma_f^{Pu}(T)$  and  $\sigma_c^{Au}(T)$  are the fission cross section of  $^{239}Pu$  and the capture cross section of  $^{197}Au$  for neutrons of the Maxwellian spectrum with a temperature of 22°C;  $P_{Au}$ ,  $P_{Pu}$ ,  $A_{Au}$ ,  $A_{Pu}$ ,  $B_{Au}$ , and  $B_{Pu}$  are the weights, atomic weights, and bond energies for Au and  $^{239}Pu$ ; and  $\delta$  is a factor depending on the geometry of the  $^{239}Pu$  and Au specimens.

From the above expression it follows that the reliability of the measurements is greater when  $\alpha(E) = \alpha(T)$ , because then the errors in the second square brackets are eliminated.

The data on  $\alpha$  and  $\sigma_f$  were averaged over the recommended energy ranges and are listed in Table 1. For  $\alpha$ , when  $E_n < 6$  keV we give data for a specimen with  $\bar{n} = 4.2 \cdot 10^{20}$  nuclei/cm<sup>2</sup>, and when  $E_n > 6$  keV, for a specimen with  $\bar{n} = 4.9 \cdot 10^{21}$  nuclei/cm<sup>2</sup>.

Table 1 gives the errors determined by the rms scatter of the data and by the normalization. The total error was determined as the square root of the sum of the squares of these errors. The correction for the resolution of the spectrometer for  $\alpha$  is not more than 4% for neutron energies of 1-30 keV.

In Fig. 1 our own results are compared with those of other authors obtained since 1972 [4-8]. The results of the more recent work were analyzed by Konshin and Sowerby [9] and are considered together in the form of the data averaged by these authors.

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# EVOLUTION OF HYDROGEN FROM PROTON-IRRADIATED CONSTRUCTION MATERIALS

Yu. P. Vasin, A. G. Zaluzhnyi,  
D. M. Skorov, and O. M. Storozhuk

UDC 621.039.531.37

It is well known that under the influence of neutron irradiation austenitic steels and nickel-base alloys change their physical and mechanical properties. Thus, after neutron irradiation at high temperatures, swelling and embrittlement usually take place in these materials. There are several hypotheses which have been proposed to explain this phenomenon. For a complete understanding of the effects of neutron irradiation of such materials it is certainly essential to study the behavior of the gaseous products appearing as a result of  $(n, \alpha)$  and  $(n, p)$  nuclear reactions as well.

We studied the kinetic characteristics of hydrogen evolution from nickel and OKh16N15M3B steel while heating the samples uniformly at  $7^\circ\text{C}/\text{min}$ . The samples were taken in the form of foils  $\sim 100 \mu$  thick and were saturated with hydrogen in an ILU-100 magnetic mass-separator using a current density of approximately  $1 \mu\text{A}/\text{cm}^2$ ; the dose of 70 keV protons was  $3 \cdot 10^{16}$  particles/ $\text{cm}^2$ . During irradiation the vacuum was held at  $10^{-7}$  mm Hg in the apparatus. The samples were surrounded with a system of screens cooled with liquid nitrogen.

Saturation of the samples with hydrogen was also carried out in a cyclotron (Institute of Atomic Energy) by irradiation with 20-MeV protons. Between the proton source and the samples undergoing saturation was a rotating disk carrying specially chosen filters, so as to ensure a uniform distribution of the hydrogen over the sample volume. During the saturation period the sample temperature was monitored by means of thermal (heat-sensitive) paints; it never exceeded  $70^\circ\text{C}$  (the samples were subjected to intensive cooling). The calculated hydrogen concentration in the samples was  $10^{-2}$  at. %.

In order to study the kinetic characteristics of hydrogen evolution from the materials in the course of uniform heating at  $7^\circ\text{C}/\text{min}$ , we used the apparatus described in [1]. The working space of the system was evacuated with two pumps (getter-ion and electrical-discharge types) at a practically constant velocity. The change in hydrogen partial pressure taking place in the working space was monitored with an IPDO-1 omegatron. The experimental curves obtained by this method characterize the relative rates of gas evolution from the materials in the course of heating. Unfortunately such methods fail to determine the actual amount of gas evolved in the annealing period.

On studying samples saturated with hydrogen in the magnetic separator ( $E = 70 \text{ keV}$ ) at room temperature, no evolution of hydrogen was detected when the samples were heated to  $950^\circ\text{C}$ . The hydrogen introduced into the test materials in this way is apparently not retained, even at room temperature, at which the irradiated samples were held for three days before measuring the gas evolution. Many authors (especially Rossin [2]) have commented that materials containing hydrogen easily lose it, even at room temperature. The high mobility of the hydrogen atoms is responsible for the ease with which the gas dissolved in the metal lattice is released. Only hydrogen "fixed" in some way remains in the sample. However, in the present case the hydrogen atoms were introduced into a surface layer  $\sim 7000 \text{ \AA}$  thick, which increased the probability of hydrogen evolution and opposed the fixing of hydrogen in "traps" such as dislocations, phase boundaries, and so forth.

If, however, the hydrogen is introduced by irradiating the samples with high-energy protons, a greater defect concentration is created, and (in the case of reasonably thin samples) any depth of hydrogen penetration may occur. Uniform heating of materials saturated with hydrogen in a cyclotron (20-MeV proton

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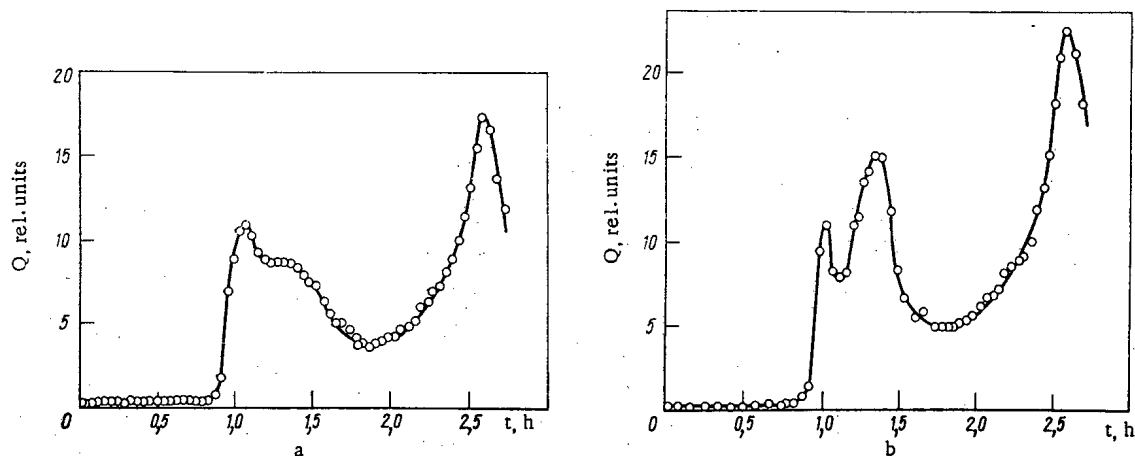


Fig. 1. Curves representing the evolution of hydrogen from OKh16N15M3B stainless steel (a) and nickel (b) samples saturated with hydrogen in a cyclotron ( $10^{-9}$  at. %) during the uniform heating of the samples at 7 degC/min, the working space being evacuated at a constant rate.

energy) led to the evolution of the gas, whereas the heating of samples irradiated with 70-keV protons did not.

Typical curves representing the evolution of hydrogen from cyclotron-irradiated samples of OKh16N-15M3B steel and nickel on heating at 7 degC/min are shown in Fig. 1. In order to determine the reproducibility of the results, at least two hydrogen-evolution curves were obtained for each sample material. The difference from one sample to another amounted to a slight displacement of the temperature peaks ( $\sim 20^\circ\text{C}$ ), without any change in the character of gas evolution. The curves representing the evolution of hydrogen from the OKh16N15M3B and nickel samples are characterized by three peaks in the temperature ranges 430–450, 550–600, and 1050–1100°C. No gas evolution occurred under 300°C. Since the diffusion mobility of hydrogen is considerable even at room temperature [2], the absence of hydrogen evolution at temperatures below 300°C indicates that the hydrogen in solid solution had already left the samples at room temperature. The hydrogen-saturated samples had been stored for 10 days. This conclusion is confirmed by the absence of gas evolution from samples saturated with hydrogen in the ILU-100 apparatus. The peaks on the evolution curve above 300°C evidently characterize the evolution of hydrogen from "traps" in the form of microcracks, pores, dislocations, interfaces, gaseous impurity atoms, and so on [3]. The disposition of the peaks on the gas-evolution curve corresponds to the activation energy of the processes involved in the "separation" of the gas atoms from the dislocations, interfaces, and so forth, and their subsequent diffusion to the sample surface. In the presence of gas porosity, gas evolution may occur in the course of its redissolution from gas bubbles, and also as a result of the migration of the bubbles themselves. The results of many authors indirectly confirm the view to the effect that microscopic discontinuities in the material are filled with hydrogen [3, 4], while autoradiographic investigations [5] directly confirm this proposition. Final elucidation and analysis of the mechanisms underlying hydrogen evolution from materials subjected to uniform heating await further research.

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# CROSS SECTION FOR THE FISSION OF $^{240}\text{Pu}$ BY NEUTRONS FROM A NUCLEAR EXPLOSION

É. F. Fomushkin, E. K. Gutnikova,  
G. F. Novoselov, and V. I. Panin

UDC 539.173.4:621.039.9

The use of neutrons from a nuclear explosion for physics research such as the measurement of fission cross sections of various isotopes by the time of flight method has a number of advantages over other pulsed neutron sources [1].

In measurements reported in [2] fission fragments were recorded by semiconductor detectors. The processing and recording of the current signal from the detector requires complex electronic equipment. The system requires careful shielding against scattered neutrons and gamma rays and electromagnetic induction.

It is therefore expedient in making measurements with neutrons from a nuclear explosion to use polymeric film as a fission fragment detector since it is insensitive to other kinds of radiations.

In our experimental arrangement for measuring fission cross sections a film of polycarbonate with a molecular weight of 90,000 developed by the Scientific-Research Institute of Plastics was fastened to the rim of a cylinder (Fig. 1) rotating  $\sim 10^4$  rpm at the instant of the neutron pulse.

Layers of fissionable isotopes were placed in the neutron flux close to the polycarbonate film, and fission fragments passing through a narrow collimator fell on the film forming tracks. The number of tracks on the part of the film corresponding to the time interval between  $t$  and  $t + \Delta t$  is

$$\Delta n = \phi(t(E_n)) \sigma_f(E_n) \Omega N_{\text{nu}} \Delta t, \quad (1)$$

where  $\sigma_f(E_n)$  is the fission cross section,  $N_{\text{nu}}$  is the number of fissionable nuclei in the layer,  $\Omega$  is the efficiency of counting fragments determined mainly by the sizes and relative position of the layer and the collimator,  $\phi$  is the local neutron flux at time  $t$ , and  $t$  is the neutron time of flight.

The time and energy resolution in the procedure described are determined by the angular velocity of the cylinder, the flight path, and the width of the fission fragment collimator slit:

$$\Delta t/L = \Delta x/vL, \quad (2)$$

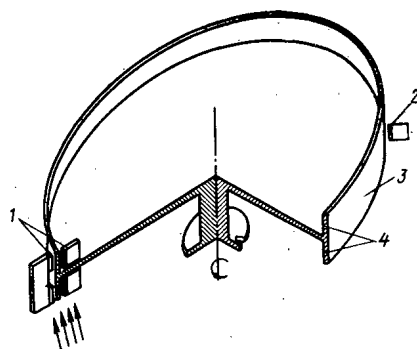
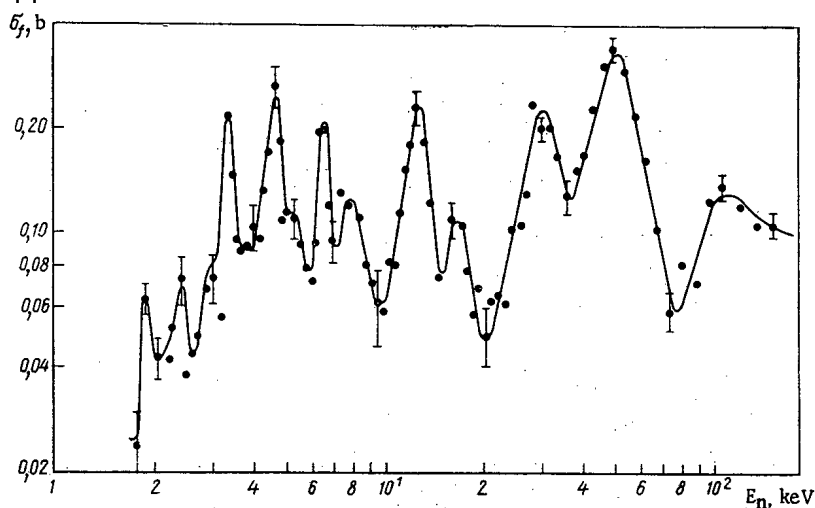


Fig. 1. Geometry of experimental arrangement: 1) collimators with layers of fissionable isotopes; 2) collimator with layers for measuring the background of scattered neutrons; 3, 4) polymeric films.

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Fig. 2. Measured values of  $^{240}\text{Pu}$  fission cross section.TABLE 1.  $^{240}\text{Pu}$  Fission Cross Section Averaged over Energy Intervals

$E_{\min}$	$E_{\max}, \text{keV}$	$\sigma_f, \text{b}$	$\Delta\sigma_f/\sigma_f, \%$
2	4	0,080	6,8
4	6	0,124	7,0
6	8	0,124	6,8
8	10	0,083	7,2
10	15	0,142	9,3
15	20	0,080	7,4
20	30	0,130	9,4
30	50	0,220	7,1
50	100	0,123	7,5
100	200	0,103	7,4

where  $L$  is the flight path,  $v$  is the linear velocity of displacement of the film relative to the collimator with the layer, and  $\Delta x$  is the collimator slit width.

In our measurements the time resolution was 12 nsec/m and the energy resolution

$$\Delta E_n/E_n \div 0.01 \sqrt{E_n (\text{keV})}. \quad (3)$$

Collimators with layers of the isotopes being studied were set up outside the direct flux to take account of the background of scattered neutrons.

We had intended to measure relative values of the  $^{240}\text{Pu}$  fission cross section by using the  $^{235}\text{U}$  fission cross

section as a reference, but because of mechanical trouble we could not obtain results with the film recording  $^{235}\text{U}$  fission products so we used data on  $^{241}\text{Pu}$  as a reference. The fission cross section of this isotope has been studied in some detail [3]; in the 1-200 keV range the cross section varies smoothly as is characteristic of isotopes with unfavorable fission thresholds.

The effective amount of fissionable material in the  $^{240}\text{Pu}$  and  $^{241}\text{Pu}$  layers, i.e., the value of  $N_{\text{nu}} \Omega$ , was measured by using a low-voltage source of 14.1-MeV neutrons. The results of White and Warner [4] were used here. The accuracy of "weighing" is 6%. The polycarbonate films were processed in a 6.25 N solution of NaOH at 42°C for 2 h. The films were scanned and tracks were counted with an MBI-9 microscope.

The results of the measurements of the cross section for  $^{240}\text{Pu}$  fission by neutrons with energies from 2 to 100 keV are shown in Fig. 2. The errors quoted are statistical. In this energy range the cross section shows a resonance structure characteristic of subbarrier fission. The positions of the first resonances ( $E_n < 4$  keV) agree with the results of Migneco and Theobald [5]. The resolution of  $\sim 10$  nsec/m is not enough to show the fine structure of the resonances due to the double-humped fission barrier. The  $^{240}\text{Pu}$  fission cross section averaged over the energy intervals is given in Table 1; the mean-square errors are cumulative.

Thus by using dielectric track detectors fission cross sections in the keV region can be measured relatively simply with neutrons from a nuclear explosion.

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PROBLEMS OF INCREASING THE SERVICE RELIABILITY  
OF THE METAL IN THE POWER EQUIPMENT OF  
NUCLEAR POWER STATIONS

L. M. Voronin and E. P. Karelin

At the end of 1974, 477 nuclear power-station units with a total power of 359 million kW were in the stage of preparation, construction, or practical service in some 30 countries. In 1975 more than 150 nuclear-power-station units with an electrical power of approximately 80 million kW are being used.

The most important factors influencing the wide introduction of nuclear power include the competitiveness of nuclear power stations at powers exceeding 100 MW and their advantages as regards effects on the environment. The absence of effluents comprising the combustion products of organic fuel (smoke, sulfur dioxide, and so on) from nuclear power stations is of great benefit to the atmosphere, which is extremely important in view of the current scale of electric power generation.

However, the specific characteristics of nuclear power stations arising from the formation of radioactive products demand special safety precautions and a high operational reliability of the equipment so as to avoid the harmful effects of radioactivity on the environment and population. Service experience confirms that the main problem in ensuring the safe operation of nuclear power stations is that of the reliability of the metal composing the equipment. This very pressing and vital theme was considered by the All-Union Scientific-Technical Conference on "Problems of increasing the service reliability of the metal in the power equipment of nuclear power stations" held in the Beloyarsk Nuclear Power Station on June 17-19, 1975.

The conference was organized by the Principal Directorate of Nuclear Power Stations attached to the Ministry of Power and Electrification of the USSR; it involved 157 specialists from 78 organizations, who presented 60 scientific papers.

A number of contributions (for example, those of the Dzerzhinskii High-Temperature Institute and the Beloyarsk Nuclear Power Station) analyzed various methods of nondestructive testing used in operational nuclear power stations and formulated the basic technical requirements for the remote-control monitoring of the equipment. Of all existing methods of defectoscopy, ultrasonic methods satisfied these requirements (nondestructive testing, high sensitivity and resolving power in the presence of background radioactive radiation, remote-control monitoring without any serious loss of time, etc.) most completely. The parallel use of active and passive ultrasonic location (sonar) facilitated the detection and classification of defects in the metal.

A number of papers were devoted to various ways of applying the ultrasonic method of defectoscopy during active service. Great interest was evoked by papers relating to installations designed and manufactured in the Beloyarsk Nuclear Power Station for the remote-control monitoring of individual types of equipment, as well as demonstrations of experimental models of several such installations. For example, there was a device for testing the annular welding seams of pipelines, comprising two ultrasonic defectoscopes of the UDM-1M type, and also a control desk with a supply unit for the remote scanning mechanism; this enables welds to be tested on both sides in all cross sections from a distance of 30 m or over. The time occupied in testing a pipe 273 mm in diameter was 6 min.

Another device facilitates long-distance monitoring of the wall thickness of a drum-separator housing, the peeling of the cladding layer, and the cracking of the surfaces and welding seams. Scanning is effected

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by means of a mechanism which moves along on electromagnetic suction feet, traveling over the surface of the drum separator in any direction and carrying search heads with it.

A special device also detects defects on the inner surface of the drums from outside without needing to remove anything inside the housing.

The Beloyarsk Nuclear Power Station staff have developed special search heads of the "GIB" type with a varying angle of incidence of the ultrasonic beam, capable of operating with all standard-production defectoscopes. Certain papers also mentioned the development of special (robot) devices of the "Magnitokhod" and Shchetochnyi Trubokhod" ("magnetic vehicle" and "brush-type tube vehicle") types for the transportation of ultrasonic test apparatus.

In combined communications of the Dzerzhinskii High-Temperature Institute, the Novovoronezh Nuclear Power Station, and the Beloyarsk Nuclear Power Station, experience in testing the state of the metal in the equipment of these power stations was correlated.

Great attention was paid at the conference to new systems and methods of testing metal, especially methods of acoustic emission. These methods were considered in papers from the Moscow Physical Engineering Institute, the Dzerzhinskii High-Temperature Institute, the Kurchatov Institute of Atomic Energy, and other organizations. The method of stress-wave emission may be extremely promising in providing continuous monitoring of the metal during service and in preventing emergency situations.

However, ways of obtaining reliable information as to the parameters of stress-wave emission signals and their dependence on the loading conditions existing in practical equipment have not yet been adequately studied, nor has the problem of recording these signals on the background of the severe noise of the working equipment and determining the location of a defect been completely solved.

There was considerable interest in a contribution from the Kurchatov Institute of Atomic Energy on the prospects of using nuclear  $\gamma$  resonance as a method of nondestructive testing. Nuclear  $\gamma$  resonance facilitates a study of the structure and phase constitution of various metals and alloys in surface layers as much as 8000 Å deep.

A method of vibrational acoustic testing based on measuring and analyzing structural (technological) noise is used with success in the Novovoronezh Nuclear Power Station. A contribution of the Dzerzhinskii High-Temperature Institute devoted to this method presented examples of damage incapable of being detected by any other methods.

The transactions of the conference show that in the last few years considerable advances have been made in organizing the service testing of metal composing the equipment of nuclear power stations. Experimental devices and installations for testing the reliability of equipment have been created and are already in use. Statistical data accumulated in the Beloyarsk and Novovoronezh Nuclear Power Stations show that the increase in the operational reliability of the equipment achieved by continuously monitoring the state of the metal and welds, together with the timely detection and elimination of defects, has a marked beneficial influence on the reliability and safety of nuclear power stations, increases the use factor of steady power production, and ultimately increases the creation of electrical power. At the same time service experience demands the creation of special systems and standard sets of instruments for the remote-control and nondestructive testing of the metal forming the basic equipment of the first circuits of nuclear power stations.

It was noted at the conference that the necessity had arisen of unifying the criteria employed for estimating the state of equipment under service conditions. It is also essential to develop the best methods of testing metals. According to published data, expenses used in the nondestructive testing of the equipment of thermal power stations contribute up to 20% of the total running expenses, while in nuclear power stations they are twice as great, so that the choice of the best testing methods is of great economic significance. It was noted at the congress that the necessity had also arisen of producing unified reference material as to the testing of metals in the equipment of nuclear power stations containing reactors of various types. Of no small importance is the problem of preparing specialists concerned with nondestructive testing methods.

The conference proved to have been extremely successful; it correlated a great deal of useful experience, and pointed the way to the further development of long-range nondestructive testing methods under service conditions; all those participating in the conference are sure that this will lead to the successful solution of the problems involved in increasing the safety and reliability of nuclear power stations.

## SECOND ALL-UNION RADIOGEOCHEMICAL CONFERENCE

R. P. Rafal'skii

The second All-Union Conference on Radioactive Elements in Rocks was held at Dushanbe on May 13-15, 1975. At this conference, which heard 40 reports, attention centered on the migration of radioactive elements in the crust in various geological processes. This problem has been most closely studied from the geological viewpoint, on the basis of field studies of various geological objects and subsequent laboratory investigations of the stony material. The physicochemical conditions of migration of uranium and thorium were also discussed on the basis of experimental and thermodynamic data. The reports dealt with the behavior of isotopes of radioactive elements in geological processes, the influence of radioactivity on the thermal conditions in the earth, and several other questions.

The laws of distribution of radioactive elements in geological formations within the USSR were analyzed in a report by A. A. Smyslov, A. I. Tugarinov, A. B. Ronov, V. M. Terent'ev, V. K. Titov, and G. M. Shor, who correlated a large amount of data on the contents and chemical states of uranium and thorium in minerals, rocks, and geological formations in various structural zones. This enabled them to establish the relations between the processes of concentration and dissemination in relation to the external factors of migration. On the basis of an analysis of the relative roles of individual geological processes (syngenetic — sedimentation and magmatism — and epigenetic — regional, contact, and hydrothermal metamorphism and epigenetic alterations due to the activity of subsurface water) in the formation of zonation in the distribution of uranium and thorium, the authors assessed the general directedness of their migration in the crust in various stages of its geological development. In particular, in deep zones of pre-Cambrian folded regions (granulitic facies) they found marked loss of uranium, and to a lesser extent thorium, from below upward as a result of regional metamorphism and ultrametamorphism. In the platform stage of development of the crust, transport of uranium in two forms (in the dissolved state and with fragments of rock-forming and accessory minerals) leads to the appearance of two fundamentally different types of radiochemical provinces.

A typical radiochemical feature of Phanerozoic folded regions is zonation manifested in a gradual weakening of migration of uranium and thorium from miogeosynclinal zones to eugeosynclinal zones.

On the basis of studies of the laws of distribution of uranium and thorium in various geological formations and of various other geological and geochemical features, R. B. Baratov, V. I. Kozyrev, and S. I. Shchukin distinguished radiogeochemical provinces in Central Asia and Yu. V. Il'inskii distinguished them in Siberia. In Siberia there are two provinces, corresponding to regions of Paleozoic and Proterozoic consolidation, and characterized by different Th/U ratios in the rocks of the most important geological formations.

A study of the uranium contents of the rocks of the Ukrainian shield revealed that the intensity of metamorphic processes and the uranium contents of the primary substrate have a marked influence on the distribution of uranium and thorium in the products of progressive regional metamorphism. The latter caused widespread extraction of uranium from the original rocks, a process which becomes more marked when the degree of metamorphism increases. Rocks of a granulitic facies are found to become progressively depleted of uranium, where its remaining principal carriers are accessory minerals. On the other hand, accumulation of uranium is associated with the final stages of metamorphism (Ya. M. Belevtsey and A. M. Zhukova).

Other reports confirmed the very important part played by metamorphic processes in the dispersion and concentration of radioactive elements. It was shown that the most reliable sign of potential ore content

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in metamorphic complexes is not high mean background concentration but the dispersion of the uranium concentration. Since rocks with high dispersion values, as a rule, are tectonically disturbed and structurally associated with broad fields of metamorphites, they can be regarded as a source of ore material. Solutions which filter through such complexes determine the possibility of mobilization and deposition not only of uranium but also of its associated components (selenium, vanadium, molybdenum, etc.); this is confirmed by the interrelatedness of the ores and metamorphic country rocks (N. P. Ermolaev).

Hydrothermal, contact, and regional metamorphism of effusive rocks causes a transition of much of the uranium in the main mass to a mobile form; this is a circumstance which favors its subsequent mobilization. In this connection regions of acid volcanism with subsequent tectonomagmatic activity must be regarded as the most promising for uranium mineralization (V. P. Kovalev, A. D. Nozhkin, A. G. Mironova, and Z. V. Malyasova). Hydrothermal metamorphism causes marked redistribution of uranium. Potassium and sodium metasomatism and argillization are accompanied by accumulation of uranium in the altered rocks, whereas formation of propylitic mineral associations leads to loss of uranium.

Secondary quartzites are characterized by low concentrations of radioactive elements, though in some cases local accumulations of uranium and thorium can be found in them (E. V. Plyushchev, L. A. Ryabova, and V. V. Shatov). High-temperature alteration of gneisses to metamorphogenic garnet-sillimanite quartzites leads to intensive loss of these elements; this demonstrates yet another probable source of ore material (A. S. Mitropol'skii, S. V. Mel'gunov, and V. P. Raevskii).

In the formation of geochemical zonation, an important role has been played by the migration of radioactive elements through systems of deep faults (Yu. V. Il'inskii). Marked redistribution of radioactive elements has been found within long-lived regional faults in shields. In the progressive stage of formation of tectonites we everywhere find loss of uranium and thorium (up to 30-60% of the original amount in the country rocks) with increasing degree of dislocational metamorphism. To the latter is attributed a crucial role in preparing rocks for ore formation for a wide range of uranium ore formations of various ages. Tectonites of the green schist series (diafluorites) are regarded as one of the main sources of uranium in the formation of hydrothermal deposits (V. K. Titov, T. V. Bilibina, A. D. Dashkova, V. K. Il'gin, D. I. Makarova, and L. Ya. Shmuraeva).

Despite the fact that the problem of the source of radioactive elements in ore deposits was not the object of special discussion at the conference, it was touched upon to some extent in many of the reports. Some of the papers emphasized the role of deep sources. A study of explosion pipes of alkaline potash rocks and xenoliths included in them revealed that there is a high content of radioactive elements in the lower crustal horizons and upper mantle and also in deep magmatic melts (E. A. Dmitriev). In the vertical cross sections of granitoid massifs the maximum uranium concentrations occur in their middle and lower parts, whereas from the upper parts uranium has been disseminated into the external zones of the intrusive chambers. As a result, the granitic magma, having given rise to the Permo-Triassic intrusives of the region, can act as a source of uranium in subsequent processes (S. M. Babakhodzhaev). Supply of uranium with endogenous fluids arriving in the water of the basins plays an important part in the formation of even sedimentary uranium deposits (M. N. Al'tgauzen). Evidence for the accumulation of uranium in the residual portions of the melts and the fluid phase in magmatic crystallization is supplied by the results of a study of the local distribution of uranium in magmatic rocks, based on the method of fragment radiography. Magmatic rock containing pore solutions is regarded as a long-lived diffusion system in which there is a continuous transition of uranium from accessories to secondary minerals (leucogenes, oxides and hydroxides of iron, chlorite, etc.). Uranium associated with these minerals is easily leached out of the rocks, so that the latter can act as a source of the metal in exogenic-epigenetic deposits (B. I. Omel'yanenko and O. P. Eliseeva).

The conditions of mobilization and deposition of uranium during recent deposit formation were analyzed by A. V. Kochenov and S. D. Rasulova, who distinguished the chief types of concentrations in recent bodies of water, and showed that recent concentrations correspond to their analogs among ancient sedimentary uranium-bearing rocks and ores. They found that evaporative concentration in soda lakes is accompanied by an increase in uranium content in proportion to the increase in the carbonate ion content. Uranium does not get into the muds rich in reducing agents, characterized by low values of Eh ( $-330$  mV). Transition of uranium from soda lake waters into solid phases requires a fall in pH; pH gradients lead to a redistribution of uranium in sedimentary strata also (V. M. Gavshin, V. A. Bobrov, B. A. Vorotnikov, N. M. Nikolaeva, and A. O. Pyalling). In the accumulation of uranium in solid bitumens and brown coals, local interactions of solutions with organic substances at their places of direct contact have probably played a part which was

at least as important as that of deposition of uranium on widespread geochemical barriers which might anyway be absent (V. F. Pen'kov and V. A. Uspenskii). Furthermore, deposition of uranium on geochemical barriers might be caused by a reduction in the redox potential, due to the presence of sulfate-reducing bacteria; this was confirmed by experimental data (E. M. Shmariovich, G. F. Agapova, and D. N. Khitarov).

A particular example of the interpretation of the conditions of formation of an ancient infiltration-epigenetic uranium mineralization demonstrated the effectiveness of the use of a combination of up-to-date geological and physical methods for this purpose (S. D. Rasulova and A. S. Stolyarov).

A significant role in the work of the conference was played by discussion of the physicochemical conditions of migration of radioactive elements. The presently known general laws of migration do not reveal significant differences in the behavior of radioactive elements in particular geological situations; this is because there are a considerable number of simultaneously acting interrelated factors in various combinations. To solve the problem we must use cybernetic models, and to construct these we must invoke both physicochemical and geological information. The former form the reference points of the model, while the latter determine the structure of their combination and the initial and boundary conditions. The universality, multidimensionality, and variety of scale of the cybernetic model enables us to solve many difficulties arising in attempts to construct universal genetic classifications based on external signs (G. B. Naumov).

Physicochemical information required for a theoretical discussion of the conditions of migration include primarily the standard thermodynamic functions of the substances and the equilibrium constants of the chemical reactions. Their determination constitutes the most important problem in the experimental investigations associated with studies of the problem of migration. A so-called simulation experiment has limited value owing to the impossibility of sufficiently strict control of its conditions, which destroys the comparability of the experimental data obtained by different authors and consequently makes it impossible to gradually accumulate data and correlate them (R. P. Rafal'skii). In recent years much work has been done on the experimental determination of the stability constants of complexes of uranium and thorium at elevated temperatures, and a critical analysis of the literature data has been made in order to select the most reliable values of the constants in standard conditions (I. L. Khodakovskii, G. B. Naumov, A. A. Nikitin, É. I. Sergeeva, O. F. Mironova, and A. P. Zhidikova). On the basis of the results of simulation experiments an attempt has also been made to classify certain igneous and metamorphic rocks with regard to their capacities to precipitate uranium from hydrothermal solutions (V. B. Koval' and V. I. Nikolaenko).

An experimental study of the formation conditions of brannerite and davidite revealed that the synthesis of these minerals in hydrothermal conditions is favored by an acid medium, whereas the action of solutions with a nearly neutral or alkaline reaction leads to the decomposition of brannerite and the formation of oxides (K. G. Korolev and G. V. Rumyantseva). Solution of primary uranium minerals and subsequent redistribution of uranium in natural conditions is promoted by their metamict decomposition; this applies primarily to coffinite (Yu. M. Dymkov and B. G. Pavlov). By means of x-ray microprobe analysis, interesting information has been obtained on the migration of radiogenic lead in natural uranium minerals. Slightly oxidized pitchblendes, regardless of their ages, are characterized by a uniform distribution of lead, but when the pitchblende is oxidized and hydrated the radiogenic lead is redistributed. Coffinites and brannerites also have nonuniform distribution of lead: They can lose a considerable amount of it. In the absence of superimposed processes, lead can migrate to distances of up to 1000  $\mu$ . These data are interesting for the estimation of the possibilities of using various uranium minerals to determine ages by the lead method (V. V. Pavshukov, L. V. Komlev, E. B. Anderson, and I. G. Smyslova). New perspectives in research on the migration of radioactive elements and determination of the age of mineralization are opened by a study of radiation-induced defects in quartz by means of electron spin resonance (A. M. Danilevich and V. V. Pavshukov).

The conference also discussed such questions as the use of data on the isotopy of radioactive elements in geological research (V. I. Malyshev; D. K. Osipov and R. S. Zhuravlev; V. V. Anan'ev, L. L. Leonova, N. A. Titaeva, and V. V. Anikina); the estimation of the radiogenic component in the thermal balance of the earth (E. A. Lyubimova, A. I. Tugarinov, N. I. Arshavskaya, and O. P. Sobornov; A. A. Smyslov and U. I. Moiseenko; Yu. P. Bulashevich and Yu. V. Khachai); and certain methodological problems in the determination of radioactive elements in rocks (O. P. Sobornov; A. L. Yakubovich and M. E. Kotsen; B. M. Moiseev and L. T. Rakov). It was emphasized that it will be necessary to create All-Union standards with respect to which radioactive elements could be determined in conformity with the requirements

of geochemistry and the analytical possibilities (O. P. Sobornov). Yu. V. Il'inskii and G. B. Kochkin discussed the laws of formation of the radiogeochemical background of rocks, and E. N. Kutsel' discussed those of subsurface waters.

In the concluding report A. I. Tugarinova touched on some crucial problems concerning the migration of radioactive elements in the crust, the origins of uranium provinces, the methodological approaches to the study of the source of uranium in ore deposits, new methods of determining the uranium contents of rocks, and certain others. As a whole the work of the conference registered marked progress in research during the last few years. There has been particular success in research on the regional zonality of radioactive elements in connection with their migration and the radiochemical regionalization of the USSR, on the behavior of radioactive elements in particular geological conditions, and on physicochemical conditions of migration which have been studied by means of up-to-date experimental and theoretical methods. The conference passed a final resolution recording success in a number of problems and listing further problems for research concerning the behavior of radioactive elements in geological processes.

# ALL-UNION CONFERENCE ON THE CHEMISTRY OF NEPTUNIUM AND PLUTONIUM

A. M. Rozen

This conference was held on March 25-27, 1975 in Leningrad and was dedicated to the 250th anniversary of the Academy of Sciences. There were over 250 participants and over 80 papers were read at two plenary sessions and six sectional meetings.

The Conference was opened by B. P. Nikol'skii who dealt with the general state of research on neptunium and plutonium in the USSR: A welcome from the Academy of Sciences was given by An. N. Nesmeyanov, and one from the State Commission on Atomic Energy by V. I. Zemlyanukhin. The historical aspects and some future prospects in the plutonium problem were considered by K. A. Petrzhak. A very interesting paper by B. Ya. Galkina, V. I. Zemlyanukhin, L. N. Lazarev, R. I. Lyubtsev, M. F. Pushlenkov, V. N. Romanovskii, and S. G. Fedorov dealt with the production of plutonium and neptunium from nuclear power-station fuel and prospects for use. This pointed out, in particular, the dominant position of water-based methods of processing power-station fuel, and improvements in these should be based on optimizing the individual operations: production of new extraction reagents, suitable diluents, flocculents, sorbents, and reagents, together with the introduction of new techniques such as the reflux process, countercurrent washing, and so on.

Over 30 out of the 80 papers dealt with the structure and coordination chemistry of neptunium and plutonium compounds. A major Russian discovery has been further extended: the observation of a new higher valency state +VII in neptunium and plutonium. A plenary paper by N. N. Krot dealt with the recently defined conditions for oxidizing neptunium to Np(VII) in neutral and weakly acid solutions, as well as with the synthesis of new compounds of Np(VII) and Pu(VII). Sectional papers dealt with the properties of new compounds of the actinides in the VII state with  $\text{EO}_4^-$  anions (M. P. Mefod'eva et al.), the electrochemical properties of Np(VII) and Pu(VII) (V. F. Peretrushkin and D. P. Alekseeva), and the behavior of Np(VII) in hydrochloric acid and carbonate media (V. P. Shilov).

There was an interesting survey of the structure and coordination behavior of the actinide cations  $\text{AnO}_2^{2+}$  and  $\text{AnO}_2^+$  in the plenary report by D. N. Suglov. Six sectional papers dealt with the electronic spectra of  $\text{NpO}_2^{2+}$  and  $\text{PuO}_2^{2+}$ , as well as with vibrational spectra, NMR spectra, x-ray spectra, and the coordination behavior of  $\text{AnO}_2^+$ .

X-ray methods have been applied to the lattice parameters of trinitrates: U(VI) — Np(VI) — Pu(VI) (Yu. F. Volkov and I. I. Kapshukov), and also with neptunium dioxide (Yu. I. Belyaev et al.). Four papers (K. V. Ilyatov, L. M. Krizhanskii, et al.) dealt with Mössbauer spectroscopy.

Instrumental methods were also widely used in other studies. In particular, it was of interest to compare the results from NMR studies of solvation and complexing for neptunyl (E. V. Iorga and V. A. Shcherbakov) with spectrophotometric study of the forms taken by hexavalent neptunium, plutonium, and americium in nitrate solutions (A. G. Rykov, V. Ya. Vasil'ev, and N. N. Andreychuk). The latter found no mononitrate complexes, and their observations would indicate that dinitrate ones are formed at once. On the other hand, Shcherbakov found mononitrate forms. Unfortunately, the papers were presented at different sections and there was no discussion.

NMR methods have been applied to the interaction of cations between quaternary ammonium salts and hexanitrate anions of tetravalent uranium, neptunium, and plutonium (B. P. Nikol'skii et al.). There is a

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covalent contribution to the interaction, although it is much less than the electrostatic one. The same method indicated that the complexes of plutonium and plutonyl nitrates with organic phosphorus compounds have a covalent component increasing in the series TBP—TBPO (A. A. Vashman et al.). Theoretical calculations were presented also for the vibrational spectra of complexes of plutonium and neptunyl nitrates with trialkyl phosphates (É. G. Teterin and I. I. Tertishnik). This line was also dealt with in several purely chemical papers. For instance, it was stated that sparingly soluble salts of anion chloride complexes of uranium, neptunium, and plutonium have been obtained,  $R_2An^{IV}Cl_6$  and  $P_2An^{VI}O_2Cl_4$  (B. P. Nikol'skii et al.), while salts of neptunium tungstic and plutonium tungstic acids have also been prepared (A. S. Saprikin and V. I. Spitsyn). V. M. Makarov, M. E. Pozharskii, and others have examined the properties of concentrated and colloidal solutions of plutonium (studies on polymers). Detailed studies have also been made on mixed oxygen-bearing plutonium hydrides (A. G. Karabash).

Solvent extraction and sorption in the chemistry of neptunium and plutonium were considered in the interesting plenary papers by V. S. Shmidt (solvent extraction) and V. I. Paramonova (sorption) as well as in nine sectional papers. In particular, data of value in simulating extraction were presented on the extraction of macroscopic concentrations of Pu(IV) by tributylphosphate in the presence of uranium and nitric acid (V. E. Vereshchagin, É. V. Renard, and V. B. Shevchenko) and also for neptunium (M. I. Konarev, V. S. Il'yshenko, et al.). Interesting information was presented on the coordination of plutonium, neptunium, and uranium by new polydentate extractants (phosphaso compounds) by D. I. Skorovarov et al. The paper by A. M. Rozen, Z. I. Nikolotova, and N. A. Kartashova showed that these polydentate compounds have some unexpected features; for instance, diphosphene dioxide with electronegative substituents show reduced extraction capacity for each of the reaction centers (phosphoryl oxygen atoms), as would be expected, whereas (against all known trends) the strength of the chelates with Pu(III) and other trivalent actinides and lanthanides increases.

Several papers in this section dealt with technological questions: stabilization of Pu(IV) and Np(V), separation of plutonium and neptunium (A. S. Solovkin, V. B. Shevchenko, et al.), reductive reextraction in a two-phase system (M. F. Pushlenkov et al.), and accelerated reductive reextraction of plutonium from an amine by introducing the reducing agent into the organic phase (V. S. Shmidt et al.).

The redox kinetics of neptunium and plutonium were considered in an outstanding plenary report by B. S. Koltunov and A. G. Rykov, and also in nine sectional papers, including the oxidation kinetics of neptunium, by persulfates, and the reaction kinetics accompanying structure change in both cations (A. G. Rykov et al.), the oxidation kinetics of Np(IV) by Pu(IV), Fe(III), and V(V), and also the reduction of Np(V) by hydrazine (V. S. Koltunov et al.). It was also reported that iron catalyzes the oxidation of Np(IV) by nitrous acid and of Np(V) by iron persulfate (I. V. Shilin et al.); redox kinetics were considered for neptunium and plutonium in nitrate solutions in the presence of ozone (G. P. Nikitina et al.) and in media containing sulfuric and phosphoric acids (I. V. Moiseev et al.). It was also reported that quasiequilibria occur in non-equilibrium systems when induced redox reactions are used with plutonium (B. P. Nikol'skii et al.), and results were given on the dissolution kinetics of neptunium dioxide (Yu. G. Mashirov et al.).

One of the meetings dealt with the radiation chemistry of solutions of neptunium and plutonium; the general state of the subject was dealt with in a paper by M. V. Vladimirova et al., while the reactivity of neptunium and plutonium ions in various valency states for water radiolysis products was considered by A. K. Pikaev et al. The reactions of neptunium in nitric acid media in the presence of much uranium were considered by B. Ya. Galkin et al., as was the oxidation of Pu(IV) in nitric acid solution in the presence of high  $\alpha$ -activities (V. Ya. Vasil'ev et al.).

Nine papers dealt with the high-temperature chemistry of neptunium and plutonium. Results were reported on the state and behavior of plutonium in a chloride and fluoride melt (V. S. Ziv and V. R. Klokman) including studies on the spectra (Yu. A. Barbanel' et al.), the interaction of plutonium with oxides (Yu. I. Rodionov, V. R. Klokman, V. F. Gorbunov, et al.), the reaction of plutonium dioxide with molybdenum trioxide (G. P. Novoselov et al.), the reactions of ternary compounds of plutonium of perovskite type with fluorine (N. P. Galkin et al.), and the separation of neptunium and plutonium from transplutonium and fission-fragment elements by chloride distillation (S. S. Travnikov et al.).

Much attention was given to the analytical chemistry of neptunium and plutonium. A plenary report by V. K. Markov and B. F. Myasoedov dealt with the state and development prospects for numerous analytical methods (gravimetric, titrimetric, polarographic, coulombometric, radiometric, spectrophotometric, x-ray fluorescence, and mass spectrometric). There were also 18 sectional papers.



The conference adopted a resolution in which it pointed out the need to expand and extend theoretical researches on the chemistry of neptunium and plutonium for aqueous and organic solvents, with wider use of modern instrumental methods and the use of quantum-mechanical calculation techniques, which should provide a high scientific performance in establishing, interpreting, and predicting the structures of neptunium and plutonium complexes. Also, it was necessary to devise syntheses for new neptunium and plutonium compounds with specified properties, including inorganic ones, organometallic ones, and coordination compounds of inorganic salts with organic ligands. Further, an intensified search should be made for new selective and stable extraction and sorption agents, with extension of research into the solvent extraction of neptunium and plutonium, and also the equilibria and kinetics of solvent-extraction processes with the object of quantitative description and prediction. Finally, there should be further research on the kinetics of redox processes, efficient means of stabilizing appropriate valency forms of neptunium and plutonium, further studies in the radiation chemistry of neptunium and plutonium, improved precision and rapidity in analysis method and extended research in the high-temperature chemistry of these elements.

Particular attention in the resolution was devoted to the need to use modern mathematical methods and computing techniques for data processing, predicting equilibria, describing chemical kinetics, and simulating radiochemical technology.

The conference as a whole was well organized, but the distribution of the reports by sections was not always very good. For instance, papers on the coordination chemistry of solvent extraction were separated from other topics in coordination chemistry. As there were numerous sectional papers, which sometimes contained many details of little interest, the discussions were not very vigorous. It would be better to use other methods of conducting scientific conferences, in which the sectional papers are preceded by published extended abstracts, while the most highly qualified specialists would present critical surveys.

However, on the whole the conference showed the broad progress of the work and the numerous advances made by Soviet researchers in the chemistry of neptunium and plutonium in recent years. It would be advantageous to have regular conferences on actinide chemistry.

## THE THIRD ALL-UNION CONFERENCE ON LINEAR CHARGED-PARTICLE ACCELERATORS

I. A. Grishaev

The conference took place on June 25-27, 1975, in Kharkov University and the Kharkov Institute of Physics and Technology. One hundred and three reports were read based on problems of the physics and technology of accelerators and new methods of acceleration.

Seven sections functioned at the conference: the state, improvement and development of linear accelerators; elements of accelerators, accelerating structures, hf systems; dynamics and the formation of beams in linear accelerators; diagnostics of beam and automation of accelerators; high-energy linear accelerators, new methods of acceleration, beam requirements; special features of operation of heavy particle accelerators; possible modifications of linear electron accelerators and special features of their use in fundamental research.

In the course of the work of the conference, meetings were held in sections of the Scientific Council on Problems of Charged-Particle Accelerators, Academy of Sciences of the USSR.

At the conference there was an exchange of opinions on the future development of research and engineering programs directed at the improvement of the existing accelerators, the development of new systems, extension of the use of accelerators to related sciences and the national economy. It was noted that linear accelerators are efficient devices for investigations in conventional fields and even today may become the principal tool for radiation research and technology, including the activation analysis of auriferous ores and petroleum well logging. It is well-known that the use of linear accelerators in radiation-technological processes, per single ruble investigated gives five rubles of pure profit over five years.

The significant contribution to the improvement of the physics and technology of linear accelerators introduces operative reactors, in particular the largest scale accelerators — 2 GeV and 300 MeV in the Kharkov Physicotechnical Institute, 60 MeV in the Institute of Atomic Energy, the linear proton accelerator I-2 and the injector of the Serpukhov accelerator — on which the necessary experience is being acquired for reliability and critical operating conditions of complex systems.

Operative linear accelerators are used for solving both fundamental and applied problems. The characteristic trait of accelerators being developed and of certain operative accelerators, is their use in accelerator-storage combinations, which expand considerably the possibilities of fundamental research on the physics of high and average energies, thanks to colliding beams, meson factories, time transformation and beam monochronatization etc., and applied research on allied sciences thanks to intense fluxes of neutrons, protons, electrons,  $\gamma$  quanta, singly charged ions and synchrotron radiation in neutron spectroscopy, production of isotopes, medical — biological investigations, radiation material behavior, etc. It has been shown by universal practice that the most efficient and reliable injection system for accelerator-storage combinations can be developed on the basis of linear accelerators, which maintain in this case all the functions and possibilities of independent accelerators. Worthy of note in this connection is the approval of the experience of the Institute of Theoretical and Experimental Physics on the use of the I-2 accelerator in applied research, with conservation of the function of the injector.

The conference noted the progress in the development of a number of unique units in accelerator systems and, in particular, in research on superconductivity.

Due to the increased current in linear accelerators, great importance is attached to the further improvement of present-day methods of calculating the dynamics of the beam. Some of the reports were

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devoted to investigation of the nonlinear effects of current loading in the heterogeneous waveguide sections of accelerators, operating in both the steady-state cycle and in the stored energy cycle. The development of a self-consistent theory of current loading has permitted the solution of an important problem to be approached in a new way — optimization of the parameters of a high-energy accelerator for the purpose of increasing its efficiency and improving the phase-energy spectrum of the beam (Moscow Engineering Physics Institute and Kharkov Physicotechnical Institute).

The conference noted the great importance which has been acquired by the experimental and theoretical investigations of new types of moderating systems for linear proton accelerators and linear electron accelerators (Radiation Technical Institute and Moscow Engineering Physics Institute). In this connection, papers on the design of small-scale standing-wave electron accelerators for defectoscopy and medicine should be mentioned. The results of these papers have been the basis for the design of series-produced accelerators, developed by the Scientific-Research Institute of Experimental Physics of the Atom and the Moscow Engineering Physics Institute.

Definite successes have been achieved in the development and modernization of linear heavy-particle accelerators. Development has been completed for a working project for a linear accelerator and a technical project for a storage-buncher for an experimental meson factory assembly in the Institute of Nuclear Research, Academy of Sciences of the USSR. Installation has started of a number of the units of this complex. The multicharged ion accelerator LUMZI-10 is being modernized, for which a method has been perfected for the smooth regulation of energy and monochromatization of the beam.

Reports were presented on the development and construction of small-scale linear proton accelerators (Kharkov Physicotechnical Institute, Moscow Engineering Physics Institute, Institute of High-Energy Physics and the Radiation Technical Institute).

New results have been obtained on the excitation of large-amplitude waves in a plasma, necessary for the acceleration of charged particles, with relativistic beams. The power of these oscillations reaches 500-600 MW (Kharkov Physicotechnical Institute). The nonlinear theory of resonance and ionization methods of accelerating ions with relativistic electron beams has been refined (Kharkov Physicotechnical Institute).

Linear induction accelerators of intense beams of relativistic electrons have undergone further development (Scientific-Research Institute of Experimental Physics of the Atom, Institute of Theoretical and Experimental Physics, and the Joint Institute of Nuclear Research) and accelerators based on direct methods of acceleration and chopping of electron beams, both for self-consistent application and for the investigation of new acceleration methods (Kharkov Physicotechnical Institute, Scientific-Research Institute of Experimental Physics of the Atom, Institute of Theoretical and Experimental Physics, and the Institute of Nuclear Physics, Siberian Division, Academy of Sciences of the USSR).

The conference drafted a program of urgent investigations amongst the most important of which are the finding in principle of new methods of acceleration and the improvement of operative accelerators.

Many young specialists participated in the conference, which confirms the excellent future prospects of this trend.

## THE 2ND INTERNATIONAL SYMPOSIUM ON NUCLEAR ELECTRONICS

A. N. Sinaev

The Second International Symposium on Nuclear Electronics, organized by the Ispra research center of Euratom, was held in Stresa (Italy) on May 20-23, 1975. Approximately 250 specialists from over 20 countries and international organizations participated, and over 90 papers were presented. The symposium covered virtually all trends of nuclear electronics for high- and low-energy physics.

The Fast Electron and Measurement of Time. A paper on microchannel electron multipliers was presented by H. Petrie (France). The multipliers are based on a plate of special glass, on which are located up to  $10^6$  channels  $\sim 10 \mu$  diameter. Photomultipliers in which the system of diodes is replaced by such a microchannel plate have substantially better time characteristics than conventional multipliers: The total signal transmission time is 1 nsec, the spread of the transmission time is 100 psec, and the minimum output pulse duration is 300 psec. The new multipliers are on the market as the type HR-300. They provide the basis for new instruments whose anodes represent a matrix of  $10 \times 10$  individual elements. They can be used with advantage in hodoscopes and differential Cherenkov detectors. In addition, the multichannel multipliers may be used for image amplification, and also in high-speed high-sensitivity oscillographs. One such oscillograph, developed in France, has a bandwidth of 5 GHz, a recording speed of  $10^9$  m/sec, and a sensitivity of 7 cm/V.

Several papers dealt with time-measuring instruments, in which direct counting of the clock pulses is used in the main part of the interval, and the method of interpolation (time interval extension of roughly 100 times) at its ends. The clock pulse frequency is of the order 100 MHz, the channel width is up to 10 psec, and the measured interval up to 1 sec.

The Tomson company (France) has developed a system for analyzing fast transients for single events. It is a 20-channel system operating on the stroboscopic principle, i.e., the pulse amplitude is measured at a definite instant in each channel by means of an analog-digital converter.

The measurement range is from several picoseconds to several nanoseconds.

Analog Processing of Information. Considerable attention was paid to the theory and practice of low-noise amplifier design. There were papers on these topics from H. Miller (USA), and E. Gatti and P. Manfredi (Italy), and others. I. Lasser (USA) reported that a spectrometer with a noise level below 100 eV has been developed on the basis of a new method of computations. M. Goyot (France) described a hybrid wide-band low-noise charge-sensitive preamplifier for the simultaneous recording of energy and time information, using a high-capacity semiconductor detector.

Several papers dealt with the separation of charged particles into their two types on the basis of recorded signals. A paper from E. Gatti (Italy) and W. Radecky (USA) proposed a general method for optically discriminating between the two types of particle, by determining the maximum probability of the particle parameter values, received from several detectors. There was further discussion of the simple case when the detector outputs are fed to threshold circuits which can only take two discrete states, 0 and 1. A majority logic can then be used; in order to classify a particle as belonging to a given type, a minimum number of threshold circuits must be in the state 1.

A number of papers was devoted to analog electronic data-reading circuits with position-sensitive detectors. A survey of published work on this topic was presented by W. Radecky (USA). Semiconductor or

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scintillation detectors or proportional wire chambers may be used in the circuits. The information about the point of passage of the particles is obtained as the ratio between the currents or charges recorded from opposite ends of the detector. If a multielectrode detector is used each electrode can be connected to a tap on a delay line, and the point of passage of the particles is determined from the delay of the signals recorded from the ends of the line. Charge-sensitive preamplifiers are connected to the ends of the line. A space resolution of 0.5 mm is achieved.

Several authors discussed methods for analog retrieval of data from the cathodes of proportional chambers. A special zigzag delay line, developed at the Orsay (France) and Brookhaven (USA) laboratories, proved interesting. Papers were also presented on data-reading from each wire of a proportional chamber. As a rule, hybrid circuits are employed in such systems. The systems contain up to 9000 wires and have a resolution time of under 10 nsec.

Digital Processing of Information. In experimental high-energy physics there has been a sharp increase in recent years in the flow of recorded information of which only a small fraction is of interest; hence more attention is being paid to the fast filtering of such information before writing it into a computer. A survey of this topic was presented by M. Lezeren (CERN). In his view, the optimal solution is to develop special processors, designed for performing predetermined algorithms by equipment methods. The data processing speed can be increased by approximately two orders by performing a number of operations in parallel. CERN has developed a set of three such processors designed for experiments with proportional wire chambers. In the first processor points of the particle trajectory are determined, in the second the straight section of the track projection is found, and in the third, the entire trajectory is restored. A more complex processor, for computations from specific mathematical formulas, was directed by H. MacPherson (England).

A long paper from R. Hitt (USA) dealt with the development of the architecture of real time systems. The small computers employed in experimental work usually perform three functions: monitoring of the equipment operation, accumulation of experimental data, and interactive analysis of the data. While such a system is satisfactory for one experimenter, difficulties arise when the computer is used simultaneously in several experiments. More complex systems are therefore being developed; these have a distributive and hierarchical structure and contain several processors of computers with different functions and capacities. Such systems are extremely expensive. They should become more popular in the near future, however, following the appearance of cheap microprocessors and microcomputers, which are convenient for low-level utilization.

The microprocessor is a single silicon crystal, in which are contained all the nodes of the central processor. The microcomputer is a microprocessor, supplemented by crystals containing a memory and input-output circuits for connection with external circuits. A lot was said at the symposium about the great promise of microprocessors for nuclear electronics.

Development of the CAMAC Standard. Most of the designs presented to the symposium were built to the CAMAC standard. The problems of introducing this standard were discussed individually, as was also the development of controllers and driver units. A paper from D. Mack (USA) discussed the employment of the CAMAC standard in North America in 1975, where consecutive multibody systems, and also combinations of consecutive and branched systems, have begun to be widely used. Data can be transmitted to a distance of several kilometers by laser beam.

Two papers from P. Gallis (France) and L. Stanchi (Italy) were devoted to developments in body controllers based on the INTEL 8080 microprocessor. The first controller is located in the main body and occupies three stages, while the second is located in a special body along with the monitoring equipment. Such controllers may be used both in fully autonomous systems and also in systems which are computer terminals.

Of the branch drivers described at the symposium, mention may be made of the driver for the EC 1010 computer, developed in Hungary. The driver mates with the system of input-output instructions of this computer and can perform autonomous exchange of data blocks.

Extension of the Methods of Nuclear Electronics to Other Fields of Science and Technology. A survey of the development of nuclear radioelectronics over the 25 years of its existence was presented by E. Gatti (Italy). He noted the main achievements of Western scientists in the field of coincidence circuits, amplitude analyzers, nanosecond electronics, and low-noise amplification, etc. In Gatti's view, the science of nuclear electronics can now in essence satisfy all the requirements of physics experimentation, and the

only problem is to devise more complex systems. Further developments in nuclear electronics are dependent on the appearance of new types of radiation detector. There is thus a trend at present towards extending the methods of nuclear electronics to other fields of science and technology. The methods of low-noise amplification and spectrometric processing of signals have proved extremely promising in a number of fields.

Simultaneously with the symposium, an exhibition of new physics research equipment was held, in which the main firms of the Western countries participated. A major part of the equipment was built to the CAMAC standard. The proceedings of the symposium are to be published by Euratom.

# PRINCIPAL RESULTS OF THE APRIL SESSION OF THE INTERNATIONAL COMMISSION ON RADIOLOGICAL PROTECTION (ICRP)

A. A. Moiseev

The regular session of the ICRP took place in Brighton on April 7-17, 1975. More than 60 scientists working in the fields of radiation protection, ionizing radiation dosimetry, radiobiology, genetics, and radiation medicine participated in the work of the Principal Commission of the ICRP and its four committees.

The main attention at the session was paid to consideration of a plan of ICRP recommendations on the permissible levels of radiation which, it is assumed, will be published in 1977 and will replace the old recommendations (Publication No. 9, 1966). The new recommendations, based on the concept of nonthreshold action of ionizing radiations and a linear relation between dose and biological effect, will include a new and simplified system of permissible radiation levels (dose limits) and also information on the biological action of ionizing radiations and the risk of occurrence of somatic — stochastic and genetic effects in man. In considering this document, various data were discussed in detail, concerning the effects of the extension factor and the linear loss of energy on the result of the generic and somatic effects, radiosensitivity of the human embryo and fetus, the action of certain radiation factors, metabolism of various radionuclides, etc. Great interest was created by data on the higher production of genetic effects in man, based on information concerning the dose-effect relation for specific locus mutations in mice, and also new data on the relative dependence of dominant and recessive mutations in genesis, of certain effects and the origination of chromosome aberrations in the foetal cells of man. The estimates given showed that for males, the ratio of the somatic to the genetic risk in the case of uniform total irradiation of the body amounts to 1:1 approximately (it had been assumed previously that this ratio is 3:1 approximately). For women, the somatic risk is approximately twice as high in consequence of the exceptionally high radiosensitivity of the mammary glands. The frequency of occurrence of breast cancer in the case of irradiated females will be a factor of 6-7 higher than for leukaemia. During the discussion of the plan, problems were considered of the standardization of medical x-ray radiological procedures, taking into account the individual components of the natural radiation background, the complexity of the radiation and the nonradiation risk, and also units of ionizing radiation dosage (in particular, many had spoken out against the introduction of the new units, the "gray" and the "sievert"). In taking account of the discrepancy in the estimates of the risk of occurrence of the late consequences of the effect of ionizing radiations, the Commission charged the working group of the first committee to analyze once again the data from the many years of observations on children irradiated as a result of the atomic bomb attacks on Hiroshima and Nagasaki.

In addition to the scheme for permissible levels of radiation, the Principal Commission and Committee of the ICRP discussed in detail schemes of other publications on the various problems of radiation safety. In particular, the fourth committee, in whose duties fall the development of practical recommendations, were ready to issue four new publications.

1. Radiation Safety at Uranium and Nonuranium Mines. Here, the principal radiation factors governing the risk to the health of people working in the mines are considered; the role of these factors in the shaping of the absorbed dose and the occurrence of lung cancer; permissible levels of external and internal radiation; protective measures, directed at reduction of occupational sickness; problems of the organization of medical examination for miners; methods of measuring the concentration of radon and daughter products of its decay. In the appendices, there are brief explanations of the principal physical characteristics of radon, data on the shift of equilibrium between radon and its decay products in the mine atmosphere and data on dust dispersivity in mines.

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2. Principles and Methods of Rendering First Aid in the Case of Accidental Irradiation of Workers with Radioactive Substances and Sources of Ionizing Radiations. The classification of the conditions of accidental irradiation is given, measures of a clinical and administrative nature are considered for different cases of overexposure, methods of dosimetric measurements are given and the principal measures for rendering initial medical assistance are listed.

3. Estimation of the Degree of Radiation Safety of Planned and Accidental Discharge of Radioactive Substances into the Outside Medium. This is devoted to the methodological aspects of estimating the danger of contamination of the outside medium with liquid and gaseous discharges of radioactive substances, taking account of the migration of radionuclides through the food chain. This publication can be used for estimating the population doses of radiation of the population living in the vicinity of nuclear power stations and nuclear industrial premises; calculations of the maximum permissible discharges of radioactive substances into the outside medium, the development of emergency measures, etc.

4. Recommendations for Ensuring Radiation Safety in the Storage, Utilization and Disposal of Open Radioactive Substances in Hospitals and Medical Scientific-Research Establishments.

All these publications will be issued in 1976 by Pergamon Press in English.



THE THIRD EUROPEAN CONGRESS OF THE  
INTERNATIONAL RADIOLOGICAL PROTECTION  
ASSOCIATION (IRPA)

V. N. Lystsov

The Third European Congress of the International Radiological Protection Association (IRPA) was held on May 13-16, 1975, in Amsterdam. About 500 specialists from 24 European and five non-European countries participated in it. The main theme of the Congress was the analysis of radiation safety criteria.

Thirteen sections functioned at the Congress. The principal aspect of the conference was a report summing up the data from four to seven papers, which was afterwards followed by a general discussion. A small part of the papers, including reports by A. K. Gus'kov, L. A. Il'in, and V. N. Lystsov, was accepted for complete discussion.

Extremely interesting review reports were read at the plenary sessions. V. Jakobi (Federal German Republic) considered the risk of somatic effects as a result of the action of ionizing radiations, in which the main attention was paid to stochastic effects and, first and foremost, to radiation carcinogenesis. These effects stipulate the choice of the maximum permissible dosages. The speakers presented the results of analysis of extensive statistical data on the induction of tumors in people who endured the atomic bomb attacks on Hiroshima and Nagasaki. Mortality from leukaemia reached the maximum level in 1955. The next 15 years marked an increase in the frequency of cancer of the lungs, breasts, thyroid, and salivary glands. Due to the large latent period of these forms of cancer, the frequency of sickness must be increased. It can be expected that the ratio of the frequency of these forms of cancer to the frequency of leukaemia will be equal to 5 approximately. The absolute value of the risk for all forms of tumors amounts to about  $150 \pm 50$  cases per rem, per  $10^6$  persons. This figure is related to irradiation with a high dose intensity, and for prolonged irradiation it should be reduced, as experiments on animals confirm, by a factor of five.

The results given, reported V. Jakobi, must be taken into account in the recommendations of the ICRP. In particular, the maximum permissible dose (MPD) for whole body irradiation must be set considerably lower than the MPD for the bone marrow. The critical organ concept must be replaced by an approach in which the doses for the most diverse organs and their relative radiosensitivity are taken into account. The latest experimental data show that the assumption concerning the linear nature of the risk-dosage relation for carcinogenesis should be reconsidered. Fundamental radiobiological experiments permit the assumption that with small doses, the actual magnitude of the risk proves to be considerably lower than follows from the linear hypothesis. First and foremost, if a linear dose-effect relation can be justified for densely ionizing radiation (neutron radiation, for example), then it has a clearly expressed nature for sparsely ionizing radiation. This is associated with a significant increase of the relative biological efficiency (RBE) of neutrons in the case of a dose reduction. The dose-effect relation for the risk of occurrence of leukaemia in the survivors of Nagasaki is satisfied by the following equation:  $R = \beta D^2$ , where  $\beta = 0.003$  case per  $\text{rem}^2$  per yr per  $10^6$  survivors. From this relation, we obtain for low doses a considerably lower risk of leukaemia than results from the linear hypothesis.

The dependence of the duration of the latent period of cancer disease on the dose and its intensity is another important factor. With their reduction, as experiments on animals and a series of medical statistical data for humans show, the latent period is increased and, starting with defined doses, it exceeds the normal lifetime of the organism. Thus, even with a linear theoretical dose-effect dependence, there will

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be a threshold dose below which a cancer tumor cannot occur, as its time of induction exceeds the lifetime of the individual. For large integrated doses however, even a quadratic or linear approximation gives an approximately identical risk magnitude. Thus, for a dose of 250 rem (5 rem/yr over 50 yr), the level of risk amounts to 1%. Because of this, it is necessary to strive for a reduction of the average level of irradiation of personnel.

It is well-known that one of the most carcinogenic radioactive substances is plutonium. A paper by D. Vennart (Great Britain) was devoted to the biological consequences of the buildup of  $^{239}\text{Pu}$  in the human body. He reported that now, in Great Britain, the MPDs for plutonium for insoluble particles passing through the respiratory tracts are being reconsidered with a view to reducing the norms. The previous norms were based, in the main, on the results of experiments with radium. Vennart noted the disparity of the ICRP norms for plutonium compounds of different solubility, and also the absence of data on the natural illnesses of children whose parents worked with plutonium. The large uncertainty in the magnitude of the expected effect stipulates strongly the nonuniform distribution of the energy transferred by the radiation. Normalization of the entry of plutonium is being carefully reconsidered at the present time by the second committee of the ICRP.

Reports by K. Sankaranarayanan (Holland) were devoted to estimating the level of genetic risk and, as their main consideration, the consequences of the occurrence of genetic mutations, and A. Leonard (Belgium) discussed data on the frequency of chromosome mutations in man.

Clarification of the level of biological risk will allow us to proceed to a comparison of this risk with the advantages of using some or other nuclear technology. Certain general tendencies were contemplated in the discussions. Almost all those addressed rejected as an unreal and absurd requirement that the nuclear industry was an industry with a zero risk level. The risk, created by the introduction of nuclear power generation, must be assessed not in itself but by comparison with the risk which is created by alternative methods of power generation. Nuclear power generation remains, first and foremost, one of the "purest" and safest branches of industry.

In some addresses, the necessity which has arisen in a number of western countries to restrict the influence of defined groups of the public and press on the acceptance of solutions for the development of the nuclear industry was discussed with animation.

In the contrary case, solutions will be accepted on the basis of emotional criteria. Thus, K. Holmquist claimed that now, in Sweden, the destiny of nuclear power generation is decided not by the appropriate competent members, but by a group of five or six journalists, writing on the theme of conservation of the environment.

The recommendations of the ICRP has attracted great attention for ensuring doses which are as low as justified by social and economic considerations. Attempts are made in some papers to compare the cost of reducing the total dose with the total losses. However, specific methods of calculating the magnitude of the losses and the possibilities of utilizing the results of the calculations were subjected to criticism.

In one of the sections, the relations between primary and derived standards of radiation safety were discussed in detail. It was noted that the construction of a system of derived standards inevitably required modeling of the interaction between the individual and medium.

Considerable attention was paid to problems of dosimetry, in particular personal dosimetry. Further improvement of the high-sensitivity TLD dosimeter based on  $\text{CaF}_2:\text{Dy}$  had been successfully achieved. The crystals are shielded from damaging external effects (light, moisture, acid, contamination, etc.), by a glass capsule. Average annual doses of about 100 mR can be measured with an accuracy in excess of 2%. M. Sokhrabi and K. Morgan (USA) reported on a personal dosimeter for fast neutrons, based on a polycarbonate film, subjected to electrochemical etching. The dosimeter provides a range from 1 mrad to 1000 mrad. Because of the high sensitivity the necessity is eliminated for the use of fissile material (neptunium or thorium).

A number of reports were devoted to the organization of a dosimetric service, automation of reading and storage of data. V. Konig and G. Scheele (German Democratic Republic) considered organizational measures contributing to the systematic reduction of small doses for the personnel of medical establishments.

Among others, problems of background radioactivity (especially the radioactivity of structural materials), migration of radionuclides into the environment and the decontamination of people from radioactive isotopes should be mentioned.

The Congress was conducted at a high scientific level and its information, undoubtedly, is of interest for a wide circle of specialists on radiation safety.

# ISOTOPE RATIOS AS INDICATORS OF RADIONUCLIDE SOURCES AND ENVIRONMENTAL MIGRATION PATHS

R. M. Aleksakhin

Usually, radionuclides enter environmental objects (air, water, soil, plants, and animals) and accumulate in the human body simultaneously from several sources and by different routes. For instance, artificial radionuclides accumulate in agricultural plants on account of simultaneous influx from the air and soil, while the accumulation in the soil and plants around nuclear power plants can be due to global fallout, local escapes, influx with groundwater, and so on. Therefore, to define the causes of radionuclide accumulation in environmental objects and to monitor for radiation safety it is important to differentiate sources and migration paths; for this purpose one can use isotope ratios, bearing in mind the specificity of different sources as regards composition and the considerable selectivity for radionuclide during transport along various paths.

On June 4-6, 1975, a symposium was held in Leningrad by the Radioecology Section of the Scientific Council on Radiobiology, Academy of Sciences of the USSR and results were presented on isotope-ratio methods for environmental radiation monitoring.

Isotope ratios are used in nuclear meteorology ( $^{141}\text{Ce}/^{144}\text{Ce}$ ,  $^{141}\text{Ce}/^{95}\text{Zr}$ , etc.), and these enable one to establish reasonably accurately the origins of radioactive particles and aerosols in the atmosphere. L. I. Gedeonov et al. have used the isotope ratios for cerium and zirconium to classify results from observations over many years on radioactive fallout around Leningrad; they found that these were related to particular nuclear tests. The concentration ratios of radioactive fission products in precipitation have been used by K. P. Makhon'ko et al. to calculate the transport of artificial radioactive substances of global origin from the southern hemisphere to the northern one.

The movements of  $^{90}\text{Sr}$  and  $^{137}\text{Cs}$  in soil — plant and water — soil systems are usually evaluated by calculating the ratios of these radionuclides; this reveals clearly the much higher tendency of  $^{90}\text{Sr}$  to migrate relative to  $^{137}\text{Cs}$ . Quantitative characterization has been given for the migration rate of  $^{90}\text{Sr}$  relative to  $^{137}\text{Cs}$  for various types of soil in the USSR (K. P. Makhon'ko, A. N. Silant'ev, and V. G. Grakovskii).

Artificial radionuclides entering the environment are new ingredients, whose rate of incorporation into migration cycles gradually falls on account of various reactions, which in radio ecology are called ageing, i. e., processes related to the transfer of radionuclides to soils in nonexchangeable and largely inaccessible forms. Quantitative forecasting of environmental contamination makes it desirable to estimate the fall in the rate of transfer of radionuclides from soil to plants. R. M. Aleksakhin gave data on the ageing of  $^{90}\text{Sr}$  and  $^{137}\text{Cs}$  entering the soil from the atmosphere. Use was made of the relationships of these radionuclides to their isotopic and nonisotopic carriers: stable strontium and calcium, on the one hand, and potassium on the other. Although  $^{90}\text{Sr}$  may have been a long time in the soil, it does not become less accessible to plants, or any less incorporated in biological migration cycles, whereas  $^{137}\text{Cs}$  ages appreciably and becomes largely unavailable.

G. N. Romanov et al. presented similar data for radioisotopes of zinc, iron, cobalt, manganese, strontium, and cesium, which migrate in various environmental links: soil — plants — agriculture animals. There are fairly substantial differences in the behavior of stable and radioactive isotopes of an element, which indicates fairly complex mixing processes.

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L. A. Araratyan et al. directed attention to the importance of physicochemical factors and forms of occurrence for the behavior of stable and radioactive isotopes of alkalis and alkaline earths in soils.

The  $^{234}\text{U}/^{238}\text{U}$  and  $^{228}\text{Th}/^{232}\text{Th}$  ratios have been used by N. A. Titaeva to describe the behavior of heavy-element radioisotopes (uranium, radium, and thorium) in rock — soil — water systems. The method is also useful in evaluating the entry of uranium, radium, and thorium into plants and soils and from industrial waste. The use of phosphate fertilizers has increased the influx of uranium and radium into the soil, so it appears desirable to use isotope ratios to examine modes of entry of heavy natural radionuclides into plants. The ratio of the amounts of  $^{22}\text{Na}$  of cosmic ray and human origins to stable isotopes has enabled D. G. Fleyshman to estimate sodium exchange rates in lakes, river systems, and water animals, while the ratio of  $^{137}\text{Cs}$  to the stable  $^{133}\text{Cs}$  has made possible a more accurate study of radioactive contamination in fish.

The symposium showed that the isotope-ratio method has considerable scope for radiation monitoring of the environment, and it sets out the lines for future studies in this area.

## EXHIBITIONS

"INVENTIONS AND BRANCH LICENSED PRODUCTS"  
EXHIBITIONS IN THE "ATOMIC ENERGY" PAVILION  
AT THE EXHIBITION OF ACHIEVEMENTS OF THE  
NATIONAL ECONOMY OF THE USSR

B. A. Sokolov and E. A. Strel'nikov

Nuclear power has now become a lively field of commercial activity. Many contracts have been concluded for the construction of nuclear power stations and the presentation of technical documentation, information, and experience in licensing matters; measures have been taken to publicize the commercial objects of nuclear power and to elucidate the business conditions relating to items of nuclear technology. One of the most important measures is the exhibition organized in May of this year regarding "Inventions and branch licensed products" organized by the State Committee of Atomic Energy of the USSR with the cooperation of the All-Union External-Trade corporations "Litsenzintorg" and "Tekhnabeksport."

The exhibition acquaints visitors with exhibits destined for the popular economy of the country and for commercial realization through the external-trade organizations of the USSR, which clearly demonstrate the high scientific-technical level of Soviet nuclear science and technology.

Power-reactor construction is exemplified by models of the VVER-440, RBMK-1000, BN-350, and BN-600 reactors. Of particular interest are the fast reactors, the basis of future nuclear power, and work on their commercial realization in the form of complete supplies of equipment, construction, or the sale of licenses is extremely promising.

The acceleration-technology section contains a small-scale cyclotron (the MGTs) and linear accelerators of the LUE10-2D and Elektron-3M types developed by the D. V. Efremov Scientific-Research Institute of Electron-Physical Apparatus. The compact isochronous cyclotron, incorporating energy regulation, is intended for producing short-life isotopes widely employed in biology and medicine, for the activation analysis of various substances, and also for nuclear-physics research. The cyclotron constitutes an export item; its high scientific-technical level has been noted by specialists in other countries.

Radioisotope technology and instrument-making widely employed in various fields of the popular economy are represented by the two-channel x-radiometric analyzer FRAD-1 (All-Union Scientific-Research Institute of Radiation Technology), the multiple-counter radiometric apparatus "Mustang" (All-Union Scientific-Research Institute of Chemical Technology), and a number of instruments and devices produced by the Riga Scientific-Research Institute of Radioisotope Instrument Making.

The creation of special apparatus for research in atomic technology frequently involves the solution of many technical problems and queries in contiguous fields. At the exhibition acquaintance may be made with a method of producing artificial diamonds in the form of films developed in the I. V. Kurchatov Institute of Atomic Energy. This method is the first of its kind in the world and has been patented in leading foreign countries. A diamond tool for drilling holes, including those of the curvilinear type, in such materials as glass, quartz, ferrite, ceramics, and others is exhibited by the Sukhum Physicotechnical Institute.

Apart from the exhibits mentioned, the exhibition offers an opportunity of seeing a small-scale ionization chamber for recording neutron and  $\gamma$  radiations in the active zone of a reactor, working under the influence of high temperatures and powerful radiation sources, apparatus for detecting faulty cassettes in

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a reactor cooled with liquid metal, desalination installations, welding equipment, and other items of particular interest to specialists in the field of nuclear power and other branches of industry.

The exhibition will promote the wider exploitation of the advances of modern atomic science and technology in the popular economy of the country, contacts with commercial circles in foreign countries, and the commercial realization of the scientific-technical achievements of the corresponding departments; it will remain active until November, 1975.

## NEW INSTRUMENTS AND TECHNOLOGICAL PROCESSES

LBK  $\beta$  POTASSIUM CONCENTRATION METER

L. V. Matveev, O. G. Mikhailov,  
and E. A. Strel'chenko

The LBK  $\beta$  potassium concentration meter has been developed at the All-Union Scientific-Research Institute of Radiation Engineering for rapid determination of the content of potassium chloride, potassium, or its oxide in liquid and loose samples of potassium salts, raw materials, potassium products, and potassium fertilizers in laboratory conditions. The instrument can be used as an input transducer for automatic process control.

The instrument operation is based on measuring the emission of natural  $\beta$  radiation of  $^{40}\text{K}$  whose intensity is proportional to the potassium content in the measured product. SBM-19 and SBT-10 gas-discharge counters are used as detectors. Measurements are carried out with the aid of one of three detectors of different geometries ( $2\pi$ ,  $4\pi$ , and cylindrical) and an electronic digital system which counts the number of detector pulses for a given time. The results are displayed in percent of potassium chloride, potassium, or potassium oxide concentration. Instrument calibration consists in setting the measuring time and resetting the scaler. Calibration data are calculated from the results of measurement of the average pulse count rate from a detector filled with pure potassium chloride salt taking into account the background count rate.

The instrument can operate at temperatures from 10 to 35°C and in a relative humidity of up to 80%. The instrument operates on a 220-V, 50-Hz line voltage. The instrument is provided with a system for checking the entire measuring channel and each counter separately.

One of the three detectors is used depending on the shape and volume of the tested product: the  $2\pi$  detector is used to measure dry and moist samples up to 300 cm<sup>3</sup> in volume, the  $4\pi$  detector is employed for liquid and loose samples with a volume up to 50 cm<sup>3</sup>, the cylindrical detector can measure dry loose samples up to 400 cm<sup>3</sup> in volume. The instrument is manufactured in two models: LBK-1 and LBK-1Ts. The LBK-1Ts model is fitted with the cylindrical detector only.

## Basic Specifications of the LBK

Concentration measurement range.....	0.5 to 100% KCl
Basic instrument error .....	less than 1.0% KCl
Random component of error (rms deviation)	
with cylindrical detector.....	less than 0.4% KCl
with $2\pi$ and $4\pi$ detectors.....	less than 0.5% KCl
Measuring time	
with cylindrical detector.....	less than 10 min
with $2\pi$ and $4\pi$ detectors .....	less than 12 min

Measured results are displayed in digital form on indicator lamps, as a dc voltage from 0 to 100 mV, and in a normal unit code.

The LBK instrument successfully passed official acceptance tests, has been entered into the State List of measuring instruments, and has been recommended in GOST 4568-73 "Potassium Chloride" as a

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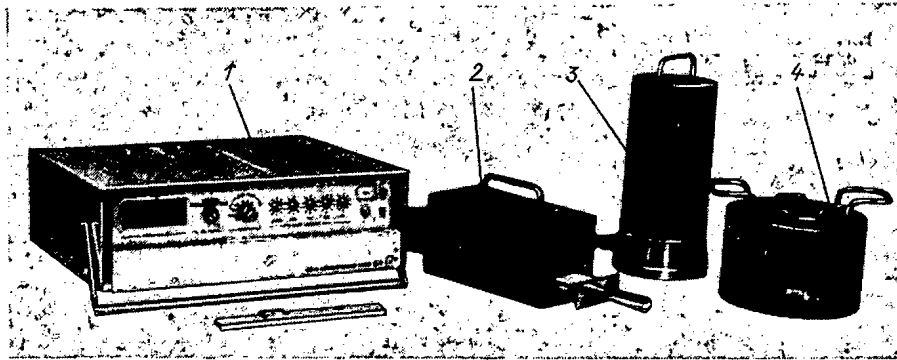


Fig. 1. LBK-1  $\beta$  potassium concentration meter: 1) electronic unit; 2)  $2\pi$  detector; 3) cylindrical detector; 4)  $4\pi$  detector.

basic measuring tool for testing the quality of potassium fertilizers. Mass production of the instrument is planned for 1976.

# A NEW METHOD OF REGENERATION OF TRAPS FOR THE PURIFICATION OF ALKALI-METAL COOLANTS

L. G. Volchkov and F. A. Kozlov

Fast reactors with sodium coolant are one of the most promising trends of nuclear power generation. Demonstration facilities EVR-II, BN-350, Phoenix, and PFR, are operating in the USA, USSR, France, and Great Britain. Intensive development is taking place in Japan, Italy, and the Federal German Republic.

The main equipment used for the purification of sodium is the continuous-flow cold trap. The maximum capacity of the cold traps is 44-47 wt. % of sodium oxide [1-3] and up to 26% of the products of interaction between sodium and water [2-3]. Filled traps must be changed or regenerated, i.e., their operating efficiency must be restored by reviving the accumulated impurities.

At present [2, 3] a distillation-gas method of regeneration is used: The trap is heated up to 350-400°C and hydrogen is removed by evacuation; then, at a temperature of 500-550°C, 80-90% of the sodium is driven off by vacuum distillation; the residues of sodium and the impurities are dissolved in alcohol and water, the solution is removed, the trap is dried out and again assembled into the facility.

The drawbacks of this method are: length of the process (regeneration of a trap with a volume of 100 liters requires 5 to 6 days), using complicated plant, the production of a large quantity of concentrated alkali solution and the presence inside the trap of exothermic reactions.

In the Physicopower Generation Institute (Obninsk), a new method of regeneration has been developed and used on traps with volumes of 100 and 800 liters. It permits purification of a trap with a volume of up to 6000-7000 liters over 30-50 h. A very small amount of additional plant and reagents is used for this and regeneration of the trap is possible without disassembly from the facility, including also industrial reactors. The regenerated product is obtained in a significantly smaller volume. Local heating inside the trap is eliminated. The new method permits multiple purification of the trap to be carried out.

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## BOOK REVIEWS

D. Bedenig\*

## GAS-COOLED HIGH-TEMPERATURE REACTORS†

Reviewed by Yu. I. Koryakin

The problem of high-temperature reactors, their possible spheres of application, is attracting great attention at present. It is true that the abundance of publications on the range of possible application of high-temperature reactors does not always correspond to the extremely modest role which these reactors play in present-day nuclear power generation and also in nuclear power generation in the immediate future (judging by current plans and future forecasts of nuclear power station installation). This abundance of publications rather reflects the anxiety for those specific technical nuclear-physical and economic indexes which it would be desirable to have in nuclear power generation, and also at present the dream of realizing a number of power-capacious, power-technical processes (not associated with electric power generation) by means of nuclear reactors.

Nevertheless, hopes for the realization of processes associated with high-temperature reactors, one of which, the gas-cooled reactor mainly for the purpose of power generation, is the subject of a quite detailed discussion in the book being reviewed.

The information in the book is based on publications and reports from Euratom, the Federal German Republic's Research Center at Julich, and companies of the Federal German Republic working in the field of gas-cooled reactors. The book is, in fact, a monographic review which is based mainly on papers about the three high-temperature reactors AVR (Julich, Federal German Republic), Dragon (Winfrith, Great Britain), Peach Bottom (USA) and, to a lesser degree, on publications about the plans and construction of the TNTP-300 reactors at Fort St. Vrain.

This is a book about reactors and their components. General plant problems of nuclear power stations with high-temperature reactors are barely considered (with the exception of the necessary technological layouts of nuclear power stations). Thus, the book covers problems and their solutions which touch upon the primary circuit, fuel and fuel elements, their loading and recharging equipments, reactor physics and control safety.

Completed projects and the current state of development of high-temperature reactors are described in the eighth chapter. As yet, these achievements are quite small and all the reactors, installed and being installed, are listed above.

The 9th chapter, concerned with plans for the development of high-temperature gas-cooled reactors, occupies a special place. Trends for the possible utilization of the reactors are distinguished in it: with a gas-turbine in a closed cycle, for technological purposes, in work on fast neutron spectra (breeder-reactors) and in conjunction with MHD generators. The achievements in these directions are extremely alluring, as this should lead either to noticeably improved values of the technicoeconomical indexes of nuclear power stations than in the present day nuclear power stations with their light-water reactors, or to an expansion of the sphere of efficient application of reactor technology, due to its introduction into large-scale technological processes. The sense and ultimate aim is the phasing out of scarce organic fuel from these processes (metallurgical and conversion of energy carriers).

\*Federal German Republic (1972).

†Yu. I. Mityaeva (editor), Atomizdat, Moscow (1975).

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The position of the author in explaining these processes is quite simple: He writes only what he would wish to have, and he is not attempting to answer the question of how this is achieved. The main problem itself consists not only in how to obtain a high temperature of the gas at the outlet from the active zone, but also how to transfer this heat and concentrate it in a radiation-safe form before the power-generation process itself, independently of whether this is a metallurgical or a coal-gassification process.

Despite the fact that from the moment of issue of the original book several years have passed, and work on high-temperature reactors is continuing, as yet there are no satisfactory solutions to this problem.

This by no means implies that the hopes for achieving significant technicoeconomical potentials of high-temperature gas-cooled reactors have changed their importance. Confirmation of this is the size of the audience of readers of the book under review — specialists interested in the problem of high-temperature reactors.

Mărgărit Pavelescu

# NUMERICAL METHODS FOR THE CALCULATION OF NUCLEAR REACTORS\*

Reviewed by S. M. Zaritskii

The Publishing House of the Romanian Academy of Sciences has brought to light a book which, obviously, was met with interest not only in Romania but also anywhere where students and graduates study the numerous methods of calculating nuclear reactors.

The book being reviewed comprises eight chapters. The different methods of successive approximation, suitable for solving algebraic or transcendental equations (Bolzano, Newton-Rawson, et al.), are considered and compared in the 1st chapter by the rate of convergence and computer time in one step. Horner's method for finding the roots of a polynomial is also described.

In the 2nd and 3rd chapters, the elements of matrix theory are discussed, and data which is associated with reactor problems is discussed quite deeply.

The 2nd chapter considers special matrices and their properties, determinants, linear vector spaces, elementary operations on matrices, eigenvalues and vectors of symmetrical matrices, functions of symmetrical matrices (raising to a power, polynomials of matrices, the matrix exponent, the Kelly — Hamilton and Sylvester theorems), eigenvalues and vectors of asymmetrical matrices, matrix transformation and Jordan's canonical form, norms and spectral radii and matrix sequences.

The 3rd chapter is devoted to nonnegative matrices. Here, the proof of Herschgerin's theorem and consequences are deduced giving estimates of the spectral radius. The properties of separable and inseparable matrices are discussed. A detailed proof is given of the Perron — Frobenius theorem and the theorem of the spectral radius of the separable nonnegative matrix. Primitive and nonprimitive (cyclic) matrices and their properties are considered. The spectral properties of nonnegative matrices are discussed.

Exact (noniteration) methods of solving systems of heterogeneous linear equations — the exclusion methods of Gauss and Jordan — are compared according to computational costs in the 4th chapter. The solution of these systems to which finite-difference approximations of one-dimensional and multidimensional reactor equations reduce is considered specially.

Finite-difference equations are investigated in the 5th chapter: finite-difference representation of linear operators and their relation with one another, and also with the operators of differentiation and integration; analytical methods of solving finite-difference equations; solutions of linear equations in partial derivatives; convergence of solutions; matrix form of finite-difference equations.

The numerical solution of ordinary differential equations is discussed in chapter six. Here, the various methods of evaluation of single and double integrals and methods of solving ordinary differential equations with boundary and starting conditions, are compared (Euler, Runge — Kutta, and Adams' methods).

In chapter seven, iteration methods of solving systems of heterogeneous linear equations are considered, which arise for example in the finite-difference approximations of multigroup diffusion equations

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of a neutron field in multidimensional models of reactors. Here, the principles of the theory of iteration methods and the problem of iteration convergence are discussed; schemes are considered together with the theoretical foundations and proofs (conditions) of convergence of the method of simple iteration, Zeidel's method and the method of point upper relaxation, the problem of determining the optimum relaxation parameter, the application and conditions of convergence of the iteration methods mentioned for solving homogeneous systems of linear equations, approximating the problems by eigenvalues. The corresponding lumped iteration methods are considered also together with their advantages over the point method.

The last, and 8th, chapter is devoted to the numerical solution of elliptical differential equations of second order in partial derivatives. Equations of particular and general form are considered, with different conditions at the boundaries of one-dimensional and two-dimensional regions of change of variables and finite-difference approximations of these equations. An analysis is conducted of the matrices to which the finite-difference approximations reduce, the convergence of the iteration methods described in the previous chapter is proved, the choice of the relaxation parameter is discussed, the convergence of the finite-difference solution to the exact solution in the case of size reduction of the lattice pitch is also discussed. Multigroup diffusion problems concerning the critical state of multizoned and one-dimensional and two-dimensional reactors are discussed.

Despite the large volume of the information presented, not all sections of reactor mathematics are covered in the book. Certain aspects in it could have been arranged in order to make explanations in some places more concise. The present-day reader, obviously, would wish to see a complete account of the problems associated with the calculation of multidimensional reactors, a review of the large number of current methods for solving these problems. However, this problem merits an independent consideration. It should be noted also, that in books of this layout, a subject index is very desirable.

On the whole, the book reviewed is a useful training and reference textbook and can occupy a legitimate place in a series, at one pole of which are practical manuals on numerical reactor calculations, and at the other pole are fundamental monographs on the theory of matrices and computational mathematics.

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